# DEVELOPMENT AND EVALUATION OF PG/SiC CODEPOSITED COATINGS FOR ROCKET NOZZLE INSERTS. VOLUME II

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Prepared for:

Air Force Rocket Propulsion Laboratory

1973

Unclassified

AD-769 286

SECURITY CLASSIFICATION OF THIS PAGE (When Data Entered)

| REPORT DOCUMENTATION PAGE | | READ INSTRUCTIONS BEFORE COMPLETING FORM |
|---|---|---|
| 1 REPORT NUMBER<br>AFRPL-TR-73-70 Vol. II | 2. GOVT ACCESSION NO. | 3. RECIPIENT'S CATALOG NUMBER |
| 4. TITLE (and Subtitle) Development and Evaluation of PG/SiC Codeposited Coatings for Rocket Nozzle Inserts. Volume II. Vapor Deposition of Pyrolytic Graphite/Silicon Carbide Codeposited Coatings | | 5. TYPE OF REPORT & PERIOD COVERED<br>Special - Jan. 1972 to July 1973<br>6 PERFORMING ORG. REPORT NUMBER<br>46-5544.11 |
| 7 AUTHOR(s)<br>Kenneth E. Undercoffer | | 8. CONTRACT OR GRANT NUMBER(s)<br>FO4611-72-C-0047 |
| 9 PERFORMING ORGANIZATION NAME AND ADDRESS<br>Atlantic Research Corporation<br>5390 Cherokee Avenue<br>Alexandria, Va. 22314 | | 10. PROGRAM ELEMENT, PROJECT, TASK AREA & WORK UNIT NUMBERS<br>Project No. 3148<br>BPSN: 623148 |
| 11. CONTROLLING OFFICE NAME AND ADDRESS<br>Air Force Rocket Propulsion Laboratory<br>Director of Laboratories<br>Edwards, CA 93523<br>Air Force System Command, United States Air Force | | 12. REPORT DATE<br>1973<br>13. NUMBER OF PAGES<br>91 |
| 14. MONITORING AGENCY NAME & ADDRESS(if different from Controlling Office)<br>Commander, DCASD Baltimore<br>Building 22, Fort Holabird<br>Baltimore, Maryland 21219 | | 15. SECURITY CLASS. (of this report)<br>Unclassified<br>15a. DECLASSIFICATION DOWNGRADING SCHEDULE<br>N/A |

16. DISTRIBUTION STATEMENT (of this Report)

17. DISTRIBUTION STATEMENT (of the abstract entered in Block 20, if different from Report)

18. SUPPLEMENTARY NOTES



19. KEY WORDS (Continue on reverse side if necessary and identify by block number)

Pyrolytic Graphite
Silicon Carbide
Rocket Nozzles
Vapor Deposition
Coatings

Details of illustrations in this document may be better studied on microfiche

20. ABSTRACT (Continue on reverse side if necessary and identify by block number)

Phase I - Task 1. Parameters were established to vapor deposit PG/SiC coatings on nozzle components with throat diameters up to 1.7 inches. The coatings were deposited from dilute methane and methyltrichlorosilane in rapidly flowing nitrogen at atmospheric pressure. The coatings were characterized with respect to morphology and SiC content. Coated throat liner components for 1.0-inch throat diameter subscale test nozzles and for 1.7-inch throat diameter HIPPO test nozzles were fabricated.



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## SECTION I

## INTRODUCTION

The basic technique for codepositing pyrolytic graphite silicon carbide (PG/SiC) coatings was developed by Atlantic Research prior to the onset of this program. Work performed for the AFRPL under Contract F04611-71-C-0014 provided process refinement and nozzle performance data which, when added to other prior work, formed the technical basis for the process studies performed on this program.

Task I of Phase I of this program was undertaken to study the specific process parameters of interest in applying coatings to nozzle throat component of 1.7-inch diameter. Uniformity and coating reproducibility were central concerns in this effort.

## SECTION II

## OBJECTIVE

The program objective is to develop a technique for fabricating reproducible PG/SiC coatings for nozzles for tactical missiles.

## SECTION III

## SCOPE

Coating techniques were developed for the fabrication of 1.0-inch and 1.7-inch throat diameter, throat inserts and entrance approach sections using, primarily, ATJ graphite as substrate material. Coating thicknesses were nominally 100 to 150 mils. Successful test firings were completed under Phase I, Task 1, and Phase II of the parent report. A few wraparound throat inserts were fabricated in both sizes. Six pieces of 1.7-inch coated hardware were sectioned and examined to check reproducibility and homogeneity of the coatings. The accuracy of the SiC content determination was checked and its relationship to codeposit density was determined. The potential of the Quantitative Metallurgical System, QMS, in accurately measuring and defining the size, shape and degree of dispersion of the SiC particles was examined in a preliminary way. Two sources of methyl trichlorosilane (MTS) and $SiCl_4$ were checked as reactant materials for SiC formation.

## SECTION IV

## CONCLUSION

Conditions were established for coating 1.0- and 1.7-inch throat diameter, throat inserts and entrance approach sections with SiC concentrations of approximately 20 percent, coating thicknesses up to 150 mils, and with a good dispersion of fine SiC particles. Good reproducibility was demonstrated. The established coating parameters for the test fired components are contained in the respective tables.

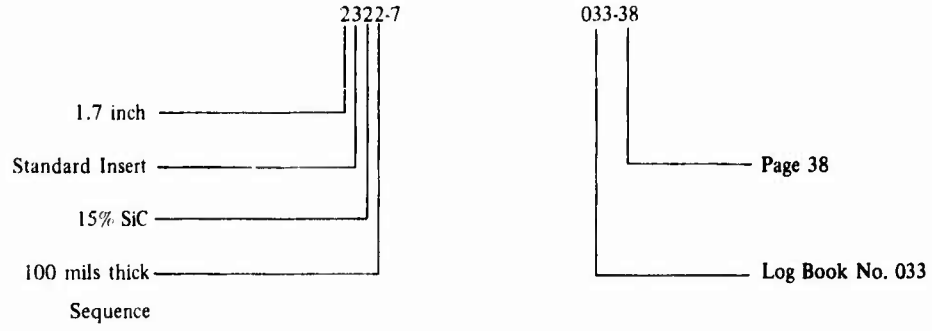
## SECTION V

## GENERAL PROCEDURES

### 1. COATING PROCEDURE

The graphite component to be coated is placed inside a graphite canister with graphite jigging in such a way that only the surface to be coated is in contact with the reaction gases. The canister is cylindrical in shape and has an inlet and an outlet graphite tube leading in and out through its front and back ends, respectively. This canister is placed inside of a horizontal carbon tube resistance furnace. The reactant gases are highly diluted with nitrogen and are maintained in the furnace under one atmosphere pressure. The reactant gases are introduced through a one-hole, water cooled, copper injector which is sketched in Figure 1. The injector spacing is defined as the distance in inches from the tip of the injector to the throat or to the exit end if an extrance approach section. The liquid methyltrichlorosilane is contained in a stainless steel upright cylindrical container with a Viton gasketed cover as shown in Figure 2. The container is immersed in a water bath and held at 90°F, while nitrogen gas is bubbled through it. Saturation, or near saturation, of the nitrogen with MTS is assumed. This gas is then further diluted with $N_2$ and methane is added. This reactant gas mixture is introduced through the injector. A small amount of nitrogen is also introduced through the annulus between the entrance graphite tube and the water cooled copper injector to prevent backflow in that area. Temperature is monitored with a Leeds & Northrup Model 8632F optical pyrometer, sighted through a hole in the graphite heater tube onto the O.D. of the canister. The calibration of the 4-inch Pereny furnace has shown a maximum difference of 40°F between this site point and the coating surface under coating conditions.

A coating index system was devised to define the type of part fabricated and the major coating characteristics. This is shown in Table I. For cataloging purposes, the part shapes were given sequential numbers based on the first 2 digits only. A second series of numbers denoted the deposition log book run number. An example of the system as used in this report is:



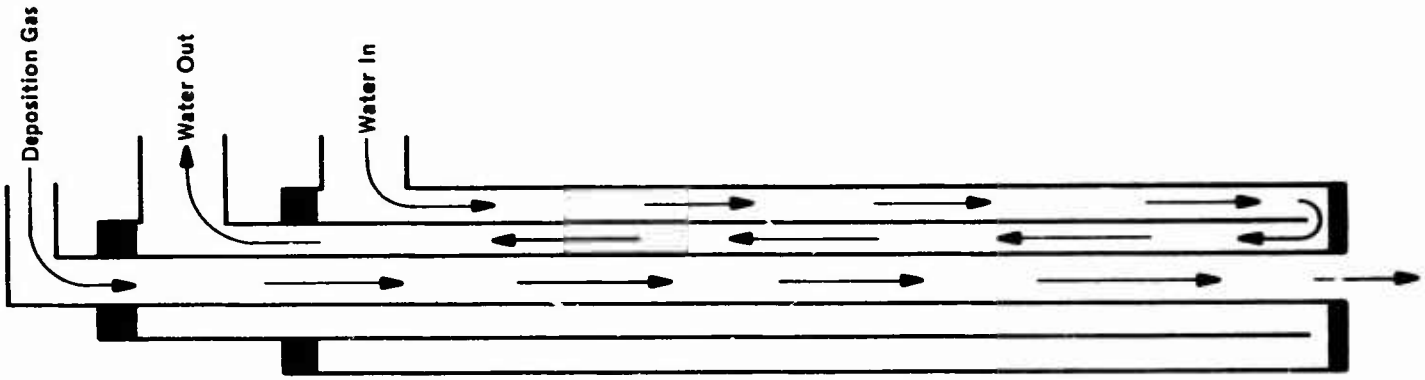



Figure 1. Water-Cooled Copper Injector.

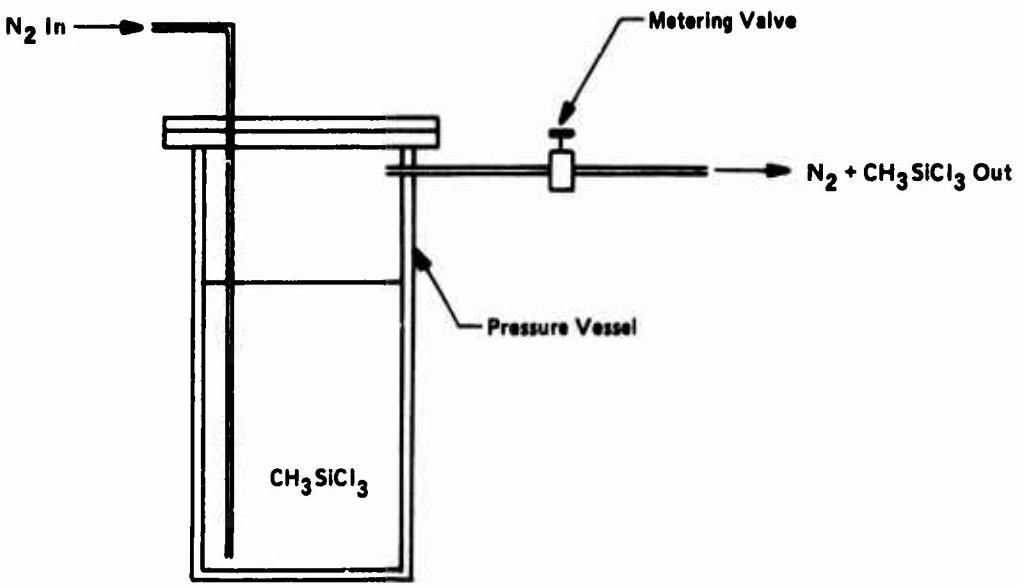



Figure 2. MTS Bubbler System.

Table I. Coating Index System.

| Description of Part | Digit 1st | 2nd | 3rd | 4th |
|---|---|---|---|---|
| **Nominal Size** | | | | |
| 1.0 | 0 | | | |
| 1.5 | 1 | | | |
| 1.72 | 2 | | | |
| 2.0 | 3 | | | |
| **Shape** | | | | |
| Standard Insert | | 3 | | |
| Wraparound - Large Radius | | 4 | | |
| Wraparound - Small Radius | | 5 | | |
| Entrance Approach | | 7 | | |
| Exit Section | | 9 | | |
| **Nominal Composition (percent SiC)** | | | | |
| 0 (Pyrolytic Graphite) | | | 0 | |
| 8 | | | 1 | |
| 15 | | | 2 | |
| 25 | | | 3 | |
| Graded Coating | | | 5 | |
| Metal Carbide (Hf, Zr) | | | 6 | |
| Ternaries (SiC + metal carbide) | | | 8 | |
| **Nominal Thickness** | | | | |
| 50 | | | | 0 |
| 100 | | | | 2 |
| 150 | | | | 4 |
| 200 | | | | 6 |
| 250 and beyond | | | | 8 |

## 2. COATING EVALUATION PROCEDURES

The insert coatings were evaluated on the basis of thickness, composition, density, and microstructure The density and composition were determined at each end on rings made available during the insert trimming operation. Microstructure was determined by polishing each end of the insert. The end coating thicknesses were also determined on the polished ends by using a filar microscope and allowing for the angle between the polished surface and the coating surface. The coating thickness at the throat was determined by measurement of the throat diameter, using calipers, before and after coating.

Density was determined on small specimens by taking a short segment of each ring and using a sink-float method with density fluids 0.05 g/cc apart at the start of the program and 0.025 g/cc apart later in the program. Composition was determined by ashing at 1,900°F in air and weighing the codeposit sample and the final $SiO_2$ product. During the program, it was discovered that weight equilibrium was not being achieved in this gravimetric analytical process and the ashing method was abandoned in favor of a density/SiC content relationship as described in Section 13.0.

The polished inserts were examined on a Zeiss inverted standard metal microscope with magnifications of 40X, 125X, 400X and 1000X. A light polarizer was used to give contrast between the PG matrix and the SiC crystals. In some cases where maximum contrast is needed, such as QMS analysis, the sample was electrolytically etched in a 5 percent KOH solution to preferentially oxidize the PG phase thereby darkening the matrix and enabling better observation of the SiC. Determination of PG cone angles and SiC crystal diameters were made by the use of angular and linear reticules placed in the microscope optics which superimposes these scales on the observed material.

A code system was devised for identification of the various types of microstructures encountered in the program. This code system, which is shown in Table II, is based primarily on SiC crystal size and degree of dispersion. The ends of the inserts were made ready for microstructural observation by standard metallographic grinding and polishing techniques. Specimens up to 4 inches O.D. were subjected to grinding on a series of silicon carbide abrasive paper. Grits No. 240, 320, 400 and 600 are normally used. The inserts were ground on a 15-micron diamond disc which is adhesive backed and mounted on an 8-inch diameter bronze polishing wheel. Water was used to carry away removed material and to lubricate the grinding surface. After grinding the insert was ready for polishing and the insert was polished on the rotating wheel. Oil and/or water was used to lubricate during the polishing process.

A 6-micron diamond polish was usually sufficient to provide clear microscopic observation. This polish could be slightly improved by use of a 1-micron diamond paste which yields a lower material removal rate.

A code system was also devised to categorize the degree of surfaced roughness of the coatings, as shown in Table III. The system is based on: (a) the average cone (surface module) diameter, (b) the size range of these cone diameters, and (c) the numbers of very large modules, or scabs present in the insert coating.

**Table II. Description of Various PG-SiC Coating Microstructural Code Types.**

| | |
|---|---|
| Group 10 | SiC phase fine, evenly dispersed, acicular, needles less than 0.035-mil diameter with an L/D ratio greater than 3/1. |
| Group 11 | Similar to Group 10 with SiC phase showing a tendency to cluster in the PG cone boundaries. |
| Group 20 | SiC phase medium sized, evenly dispersed, acicular, needles 0.035-mil to 0.105-mil diameter with an L/D ratio greater than 3/1. |
| Group 21 | Similar to Group 20 with SiC phase showing a tendency to cluster in the PG cone boundaries. |
| Group 30 | SiC phase coarse, moderately well dispersed, acicular, needles greater than 0.105-mil diameter with an L/D ratio greater than 3/1. PG growth cones usually contain occasional, intraconical, delaminations and strain lines. |
| Group 40 | SiC phase fine, well dispersed, granules less than 0.035-mil diameter. |
| Group 60 | SiC phase coarse, poorly dispersed, acicular, needles greater than 0.105-mil diameter with an L/D ratio of greater than 3/1. PG cones contain many delaminations and strain lines. |
| Group 70 | SiC phase has no definite structure. Usually consists of coarse string-like growths surrounding unalloyed PG cones. PG cones contain many delaminations and strain lines. |

**Table III. Coated Surface Condition Code.**

| | Digit | | |
|---|---|---|---|
| | 1st | 2nd | 3rd |
| Average Surface Cone Diameter | | | |
| < 10 mils | 0 | | |
| 10/20 mils | 1 | | |
| 20/40 mils | 2 | | |
| 40/80 mils | 3 | | |
| 80/160 mils | 4 | | |
| > 160 mils | 5 | | |
| Cone Diameter Range | | | |
| 0.8X* to 1.25X maximum | | 0 | |
| 0.5X to 2X maximum | | 1 | |
| 0.2X to 5X maximum | | 2 | |
| < 0.2X to > 5X | | 3 | |
| Presence of Nodules | | | |
| None | | | 0 |
| Few (< 5) | | | 1 |
| Moderate (5 to 25) | | | 2 |
| Prolific (> 25) | | | 3 |

*X = average cone diameter.

## SECTION VI

## COATING DEVELOPMENT

A total of 111 runs were made in order to fabricate sufficient throat inserts and entrance approach sections to satisfy the 1.0[1] and 2.0 inch subscale test firings at Atlantic Research and the 1.7-inch HIPPO test firings at RPL. Figure 3 shows the typical coating microstructures achieved. Considerable difficulties resulted from inaccurate SiC content determinations. It was learned during the run series that the ashing method gave low SiC contents. Consequently, some runs which were originally thought to be too low in the SiC content were eventually found to be satisfactory. This problem was corrected and all SiC contents in this report are considered to be accurate.

### 1. FABRICATION TECHNIQUE DEVELOPMENT

#### a. 1.0 Inch Nozzle Throat Inserts

Twenty-eight standard (see Figure 4) and three wraparound (see Figures 5 and 6) 1.0-inch nozzle throat insert deposition runs were conducted. The insert substrate dimensions, shown, are before coating and prior to trimming. A Pereny 4-inch carbon tube resistance furnace was utilized as the heat source. The deposition assembly configuration was similar for all 31 runs and is shown in Figure 7.

Coating process conditions are shown in Table IV and coating characterization data are shown in Table V. The microstructures were all coded recently in accordance with Table II. The process and coating evaluation data for the seven inserts that were test fired are summarized in Table VI.

All substrates, with a few exceptions as indicated in Table IV, were fabricated from ATJ graphite. In all cases the throat axis was normal to the grain direction of the graphite material used.

Substrate dimensions were recorded before and after coating to determine if any significant mismatch existed between the CTE of the coating and that of the substrate. The data are shown in Table VII. An average decrease of 0.02 percent in the substrate outside diameter was recorded indicating that the thermal expansion of the ATJ graphite in the With Grain, WG, direction is slightly lower than that of the composite coating, as expected. The substrate length showed a decrease of 0.05 percent which was completely unexpected since the measured CTE of the coating, in the ab plane, is less than that of ATJ graphite in the across grain, AG, direction. A possible explanation may be that substrate warpage occurred because of the biaxial stress fields set up by the complicated coating/substrate interface. Note that maximum stress is at the interface but the position of strain measurement is at the substrate periphery. On the other hand, the very poor consistency as indicated by the high standard deviation renders the data inconclusive.

The first five 1.0-inch inserts were fabricated under identical conditions. The injector spacing, identified as the distance between the tip of the injector and the insert throat, was 3-1/4 inches. The first of these 0332-1 was utilized for the first firing of the program, CM-1. The second 0332-2 was sectioned axially and photomicrographs

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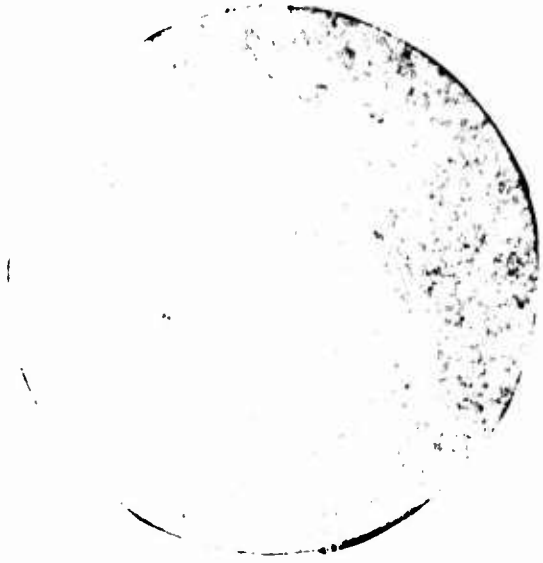
[1] The graded coatings and 20-inch coatings are not included in these 111 runs.

Group 10 Microstructure, 1260X
Entrance End View
of 1.0-Inch Throat Insert 0332-14

Group 11 Microstructure, 1260X
Exit End View
of 1.0-Inch Throat Insert 0322-16

Group 30 Microstructure, 640X
Exit End View of 1.72-Inch HIPPO
Entrance Approach Section 2742-12



Group 40 Microstructure, 640X
Axial Throat View
of 1.0-Inch Insert 0332-2

Group 20 Microstructure, 800X
Exit End View
of 1.72-Inch HIPPO Insert 2324-31

Group 21 Microstructure, 800X
Exit End View
of HIPPO Insert 2324-33

Group 60 Microstructure, 640X
Entrance End View of 1.72-Inch HIPPO
Entrance Approach Section 2742-12

Group 70 Microstructure, 1000X
Entrance End View
of 1.0-Inch Entrance Approach Section 0732-8

Figure 3. Typical Coating Microstructures.

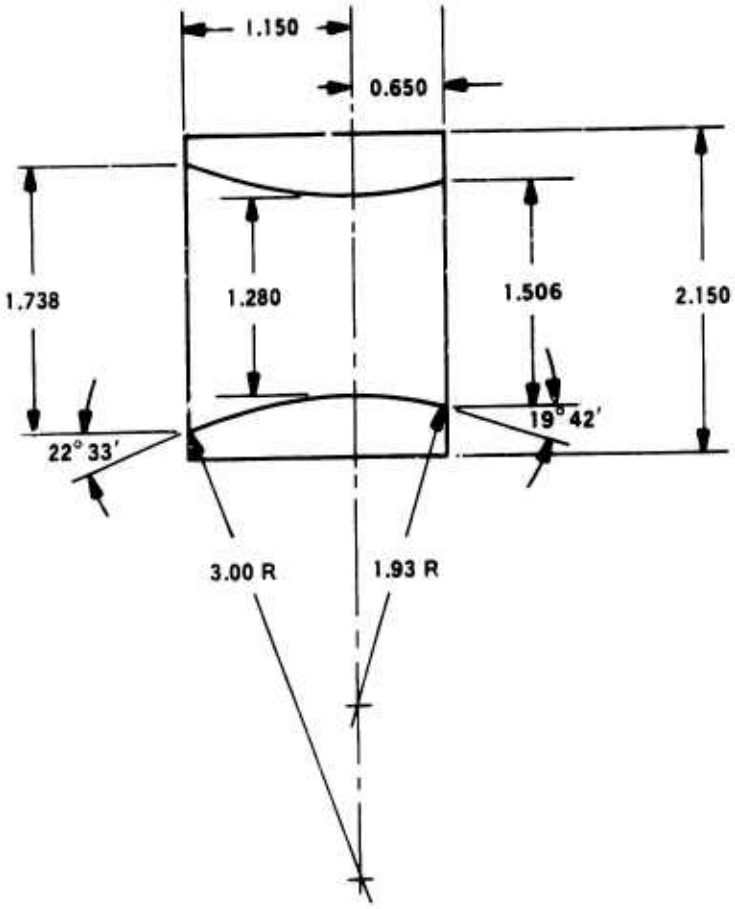



Figure 4. Standard 1.0-Inch Nozzle Throat Insert Substrate.

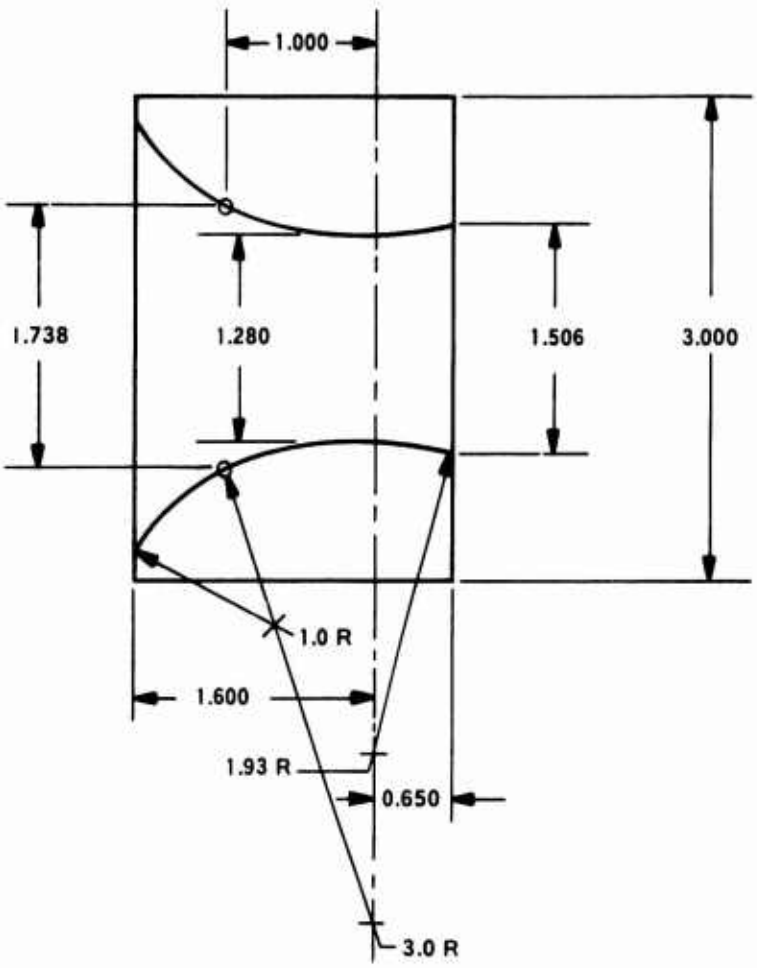



**Figure 5. 1.0-Inch Wraparound Nozzle Throat Insert Substrate (large radius).**

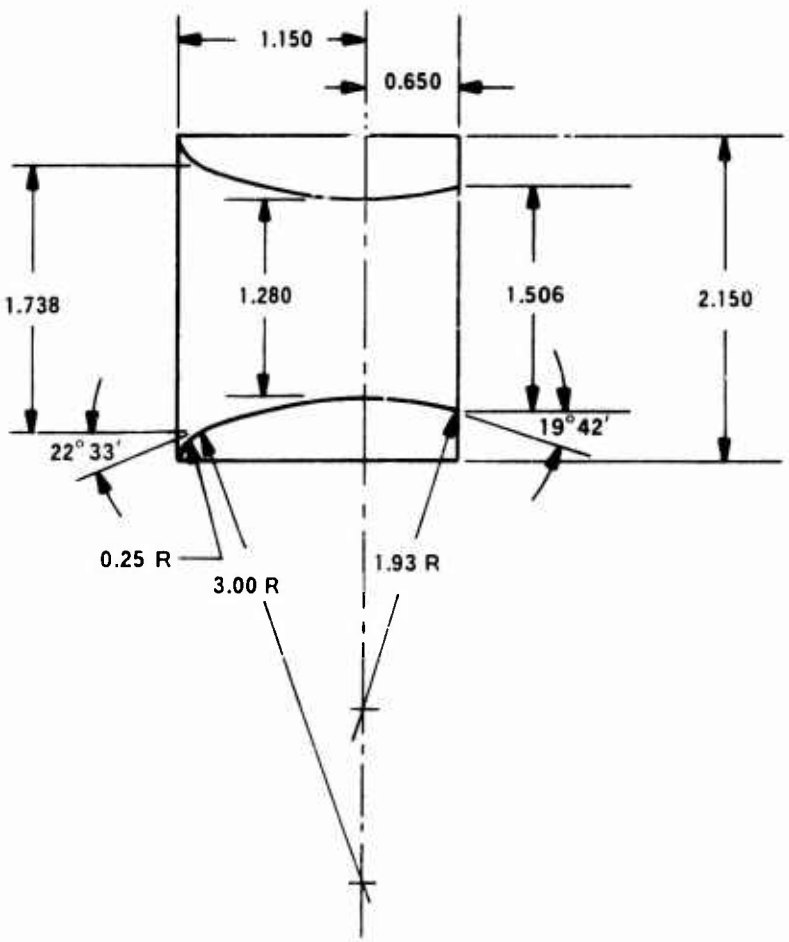



Figure 6. 1.0-Inch Wraparound Nozzle Throat Insert Substrate (small radius).

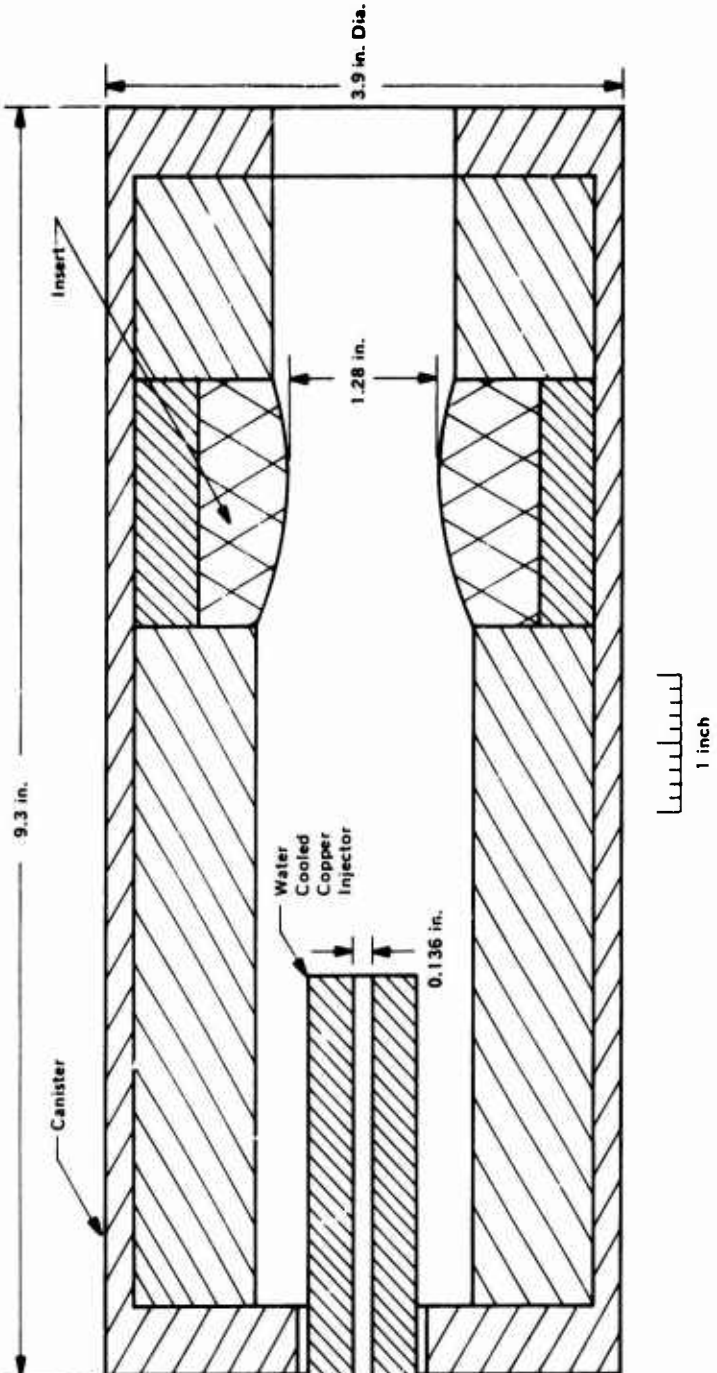



Figure 7. 1.0-Inch Subscale Throat Insert Deposition Assembly.

## Table IV. 1.0-inch Throat Insert Coating Process Conditions

| Insert Ident. No. | Deposition Run No. | Annulus $N_2$ (SCFH) | Process $N_2$ (SCFH) | Process $CH_4$ (SCFH) | Process $N_2$ to Bubbler (SCFH) | $CH_3SiCl_3$ Rate (g/hr) | Deposition Temperature (°F) | Deposition Time (hr) | Injector Spacing (in.) | Remarks |
|---|---|---|---|---|---|---|---|---|---|---|
| 0332-1 CM-1 | 162-25 | 5.0 | 140 | 2.0 | 2.5 | 54[a] | 3,200 | 6.03 | 3 1/4 | First 1.0-inch deposition run of this type. |
| 0332-2 | 162-27 | 5.0 | 140 | 2.0 | 2.5 | 54[a] | 3,200 | 6.42 | 3 1/4 | Part sectioned axially. |
| 0332-3 | 162-29 | 5.0 | 140 | 2.0 | 2.5 | 54[a] | 3,200 | 6.17 | 3 1/4 | Part sectioned axially and coating cut into four segments at entrance throat and exit planes. Data shown in Table VIII. |
| 0322-4 | 162-43 | 5.0 | 140 | 2.0 | 2.5 | 54[a] | 3,200 | 6.08 | 3 1/4 | New furnace tube installed prior to this run. Otherwise similar to 162-29. |
| 0332-5 | 162-45 | 5.0 | 140 | 2.0 | 2.5 | 54[a] | 3,200 | 6.40 | 3 1/4 | Repeat of 162-43 with increased deposition time to increase coating thickness. |
| 0322-6 | 162-48 | 5.0 | 140 | 2.0 | 2.5 | 54[a] | 3,200 | 6.40 | 3 3/4 | Injector moved back to increase coating thickness at entrance end. Controller malfunctioned during deposition. |
| 0332-7 | 162-52 | 5.0 | 140 | 2.0 | 2.5 | 54[a] | 3,200 | 7.80 | 3 3/4 | Deposition time increased to increase coating thickness. |
| 0322-8 CM-2 | 162-54 | 5.0 | 140 | 2.0 | 2.5 | 54[a] | 3,200 | 6.28 | 3 3/4 | Coating too thick during previous run. Deposition time decreased. |
| 0332-9 | 162-58 | 5.0 | 140 | 2.0 | 2.5 | 54[a] | 3,200 | 6.28 | 3 3/4 | Substrate PG precoated to facilitate removal of coating. Coating sectioned as in Run 162-29. Data shown in Table VIII. |
| 01 0332-10 CM-3 | 162-66 | 5.0 | 140 | 2.0 | 2.5 | 54[a] | 3,200 | 6.33 | 3 3/4 | Repeat of 162-58 with slightly longer deposition time to increase coating thickness. |
| 0322-11 | 162-73 | 5.0 | 30.5 | 2.0 | 4.5 | 97[a] | 3,300 | 3.00 | 3 1/4 | Deposition conditions altered in an attempt to fabricate coating with larger size SiC crystals. |
| 0332-12 CM-4 | 162-74 ATJ/A | 5.0 | 140 | 2.0 | 2.5 | 54[a] | 3,200 | 6.33 | 3 3/4 | Insert fabrication run to produce throat insert for test firing utilizing annealed ATJ as the substrate material. |
| 0332-13 | 033-5 PO-3 | 5.0 | 140 | 2.0 | 2.5 | 54[a] | 3,200 | 6.33 | 3 3/4 | Repeat of Run 162-66 utilizing PO-3 as the substrate material to determine its compatibility with PG/SiC coating. |

[a]Calculated assuming saturation of bubbler $N_2$ at 90°F.

## Tabel IV. (continued)

| Insert Ident. No. | Deposition Run No. | Annulus $N_2$ (SCFH) | Process $N_2$ (SCFH) | Process $CH_4$ (SCFH) | Process $N_2$ to Bubbler (SCFH) | $CH_3SiCl_3$ Rate (g/hr) | Deposition Temperature (°F) | Deposition Time (hr) | Injector Spacing (in.) | Remarks |
|---|---|---|---|---|---|---|---|---|---|---|
| 0332-14 | 033-8 ATJS | 5.0 | 140 | 2.0 | 2.5 | 54[a] | 3,200 | 6.33 | 3 3/4 | Repeat of Run 162-66 utilizing ATJS as the substrate material to determine its compatibility with PG/SiC coating. |
| 0322-15 | 033-21 | 5.0 | 140 | 2.0 | 2.5 | 46.2 | 3,200 | 6.33 | 3 3/4 | Conducted to re-establish deposition parameters for standard 1.0-inch insert configuration. |
| 0322-16 | 033-22 | 5.0 | 140 | 2.0 | 2.5 | 64.9 | 3,150 | 6.33 | 3 3/4 | Deposition temperature reduced 50° to determine effect on SiC crystal size. No effect was observed. |
| 0322-17 | 033-23 | 5.0 | 140 | 2.0 | 2.5 | 52.6 | 3,200 | 6.33 | 3 3/4 | Same as 033-21. |
| 0322-18 | 033-24 ATJS | 5.0 | 140 | 2.0 | 2.5 | 47.2 | 3,200 | 6.33 | 3 3/4 | Repeat of Run 033-8 utilizing ATJS as the substrate material. |
| 03XX-19 | 033-26 | 5.0 | 140 | 2.0 | 2.5 | 59.4 | 2,900 | 1.00 | 3 3/4 | Variable conditions used to determine their effect on microstructure. See Figure 8. |
| | | " | 140 | " | " | " | 3,000 | 1.00 | " | |
| | | " | 140 | " | " | " | 3,100 | 1.00 | " | |
| | | " | 140 | " | " | " | 3,200 | 1.00 | " | |
| | | " | 210 | " | " | " | 3,200 | 1.00 | " | |
| 0322-20 | 033-27 | 5.0 | 140 | 2.0 | 1.6 | 26.4 | 3,200 | 7.00 | 3 3/4 | MTS rate reduced to lower SiC concentration to determine compatibility of reduced SiC containing coating. |
| 0312-21 | 033-29 | 5.0 | 140 | 2.0 | 1.1 | 11.4 | 3,200 | 7.00 | 3 3/4 | MTS rate reduced further to lower SiC concentration. |
| 0312-22 | 033-33 ATJ/A | 5.0 | 140 | 2.0 | 1.2 | 17.7 | 3,200 | 8.00 | 3 3/4 | MTS rate increased slightly to increase SiC concentration. MTS rate erratic during deposition. |
| 0336-23 | 033-50 | 5.0 | 140 | 2.0 | 2.5 | 62.1 | 3,200 | 12.00 | 3 3/4 | Increased deposition time to obtain coating thickness of 200 mils. |
| CM-7 0332-24 (02) | 033-54 | 5.0 | 140 | 2.0 | 2.5 | 62.2 | 3,200 | 6.33 | 3 3/4 | Nozzle throat insert fabrication for test firing. Same parameters as 162-66 |
| 0332-25 | 033-56 | 5.0 | 140 | 2.0 | 2.5 | 62.4 | 3,200 | 6.13 | 3 3/4 | Run conducted to fabricate throat insert for firing 03 (CM-8). |

[a]Calculated assuming saturation of bubbler $N_2$ at 90°F.

## Table IV. (continued)

| Insert Ident. No. | Deposition Run No. | Annulus $N_2$ (SCFH) | Process $N_2$ (SCFH) | Process $CH_4$ (SCFH) | Process $N_2$ to Bubbler (SCFH) | $CH_3SiCl_3$ Rate (g/hr) | Deposition Temperature (°F) | Deposition Time (hr) | Injector Spacing (in.) | Remarks |
|---|---|---|---|---|---|---|---|---|---|---|
| 03 0332-26 CM-8 | 033-57 | 5.0 | 140 | 2.0 | 2.5 | 60.9 | 3,200 | 6.22 | 3 3/4 | Repeat of 033-56 with extended deposition time to increase coating thickness and, thereby, reduce insert throat diameter. |
| 0334-27 | 033-58 | 5.0 | 140 | 2.0 | 2.5 | 62.6 | 3,200 | 8.80 | 3 3/4 | Deposition time extended to increase coating thickness to 150 mils. |
| 0334-28 CM-9 | 033-61 | 5.0 | 140 | 2.0 | 2.5 | 56 2 | 3,200 | 9.17 | 3 3/4 | Run conducted to fabricate throat insert for firing CM-9. Deposition time longer than 033-58 to increase coating thickness. |
| 0422-1 | 033-28 | 5.0 | 140 | 2.0 | 2.5 | 55.8 | 3,200 | 6.33 | 3 3/4 | Run conducted to fabricate insert with 1.0-inch radius entrance and wraparound. |
| 0422-2 | 033-44 | 5.0 | 210 | 2.0 | 2.5 | 50.7 | 3,200 | 7.00 | 4 1/4 | Same as 033-28 utilizing annealed ATJ substrate. Injector spacing increased to increase entrance and coating thickness. |
| 0532-2 | 033-13 | 5.0 | 140 | 2.0 | 2.5 | - | 3,200 | 6.33 | 3 3/4 | Run conducted to fabricate insert with 1/4 inch by 90° entrance end wraparound. |

## Table V. 1.0-inch Throat Insert Coating Evaluation Data.

| Ident. No. | Deposition Run No. | Coated Throat Diameter (in.) | Coating Thickness (in.) | | | Density (gm/cc) | | | SiC Concentration[a] (%) | | | Appearance | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | | | Entrance | Throat | Exit | Entrance | Throat | Exit | Entrance | Throat | Exit | Microstructure | Cone Angle (degrees) | Surface Condition |
| 0332-1 CM-1 | 162-25 | 1.089 | 0.089 | 0.095 | 0.077 | 2.35/2.40 | -- | 2.30/2.35 | 30 | -- | 24 | -- | -- | -- |
| 0332-2 | 162-27 | 1.065 | 0.094 | 0.108 | 0.090 | 2.35 | -- | 2.30/2.35 | 27 | -- | 24 | Group 40 | -- | -- |
| 0332-3 | 162-29 | 1.068 | 0.059 | 0.105 | 0.072 | 2.35 | 2.35/2.40 | 2.30 | 27 | 30 | 21 | Group 10 | -- | -- |
| 0322-4 | 162-43 | 1.090 | 0.061 | 0.095 | 0.083 | 2.25/2.30 | -- | 2.25/2.30 | 18 | -- | 18 | Ent. - Group 11 Exit - Group 21 | -- | -- |
| 0332-5 | 162-45 | 1.083 | 0.056 | 0.098 | 0.095 | 2.30/2.35 | -- | 2.30/2.35 | 24 | -- | 24 | Group 21 | -- | -- |
| 0322-6 | 162-48 | 1.115 | 0.057 | 0.082 | 0.076 | 2.30 | -- | 2.30 | 21 | -- | 21 | Coating contains 1 band of Group 30 microstructure Group 21 | -- | -- |
| 0332-7 | 162-52 | 1.043 | 0.060 | 0.124 | 0104 | 2.30/2.35 | -- | 2.30 | 24 | -- | 21 | Group 10 (Almost 40) | -- | -- |
| 0322-8 CM-2 | 162-54 | 1.075 | 0.075 | 0.101 | 0.086 | 2.30 | -- | 2.25 | 21 | -- | 15 | Group 21 | -- | -- |
| 0332-9 | 162-58 | 1.094 | 0.061 | 0.095 | 0.083 | 2.35/2.40 | 2.35/2.40 | 2.35 | 30 | 30 | 27 | Group 11 | -- | -- |
| 01 0332-10 CM-3 | 162-66 | 1.077 | 0.084 | 0.101 | 0.087 | 2.35 | -- | 2.30/2.35 | 27 | -- | 24 | Ent. - Group 11 Exit - Group 21 | -- | -- |
| 0322-11 | 162-73 | 1.164 | 0.019 | 0.057 | 0.047 | 2.20/2.25 | -- | 2.30 | 12 | -- | 21 | Group 70 | -- | -- |
| 0332-12 CM-4 | 162-74 ATJ/A | 1.082 | 0.072 | 0.098 | 0.093 | 2.35 | -- | 2.35 | 27 | -- | 27 | Group 21 | -- | -- |
| 0332-13 | 033-5 PO-3 | 1.072 | 0.086 | 0.103 | 0.086 | 2.35 | -- | 2.30 | 27 | -- | 21 | Group 11 | -- | -- |
| 0332-14 | 033-8 ATJS | 1.118 | 0.068 | 0.090 | 0.077 | 2.35 | -- | 2.35 | 27 | -- | 27 | Group 10, 7-mil band of unalloyed PG at interface | -- | -- |
| 0322-15 | 033-21 | 1.138 | 0.071 | 0.099 | 0.065 | 2.25/2.30 | -- | 2.25 | 18 | -- | 14 | Group 11 | -- | -- |
| 0322-16 | 033-22 | 1.092 | 0.075 | 0.096 | 0.068 | 2.25/2.30 | -- | 2.25/2.30 | 18 | -- | 18 | Group 11 | -- | -- |
| 0322-17 | 033-23 | 1.080 | 0.082 | 0.100 | 0.070 | 2.20/2.25 | -- | 2.25/2.30 | 12 | -- | 18 | Group 60 (est.) | -- | -- |

## Table V. (continued).

| Ident. No. | Deposition Run No. | Coated Throat Diameter (in.) | Coating Thickness (in.) | | | Density (gm/cc) | | | SiC Concentration[a] (%) | | | Appearance | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | | | Entrance | Throat | Exit | Entrance | Throat | Exit | Entrance | Throat | Exit | Microstructure | Cone Angle (degrees) | Surface Condition |
| 0322-18 | 033-24 ATJS | 1.082 | 0.062 | 0.100 | 0.071 | 2.20/2.25 | -- | 2.25/2.30 | 12 | -- | 18 | Group 11 (est.) | -- | -- |
| 03XX-19 | 033-26 | 1.088 | 0.057 | 0.069 | 0.051 | | | Variable Layer Coating | | | | -- | -- | -- |
| 0322-20 | 033-27 | 1.088 | 0.075 | 0.096 | 0.071 | 2.25/2.30 | -- | 2.25 | 18 | -- | 15 | Group 11 (est.) | -- | -- |
| 0312-21 | 033-29 | 1.108 | 0.075 | 0.086 | 0.068 | 2.15/2.20 | -- | 2.20/2.25 | 3 | -- | 12 | Group 40 (est.) | -- | -- |
| 0312-22 | 033-33 ATJ/A | 1.079 | 0.097 | 0.100 | 0.079 | 2.20/2.25 | -- | 2.20/2.25 | 12 | -- | 12 | Coating contains 2 major delaminations. Group 40 | -- | -- |
| 0336-23 | 033-50 | 0.997 | 0.143 | 0.201 | 0.159 | 2.35/2.40 | -- | 2.30/2.35 | 30 | -- | 24 | First 4-6 mils of coating contains a fine delamination. Group 21 | -- | -- |
| 02 0332-24 CM-7 | 033-54 | 0.981 | 0.068 | 0.105 | 0.082 | 2.35/2.40 | -- | 2.25/2.30 | 30 | -- | 18 | Group 20 | -- | -- |
| 0332-25 | 033-56 | 0.999 | 0.060 | 0.096 | 0.072 | 2.35/2.40 | -- | 2.25/2.30 | 30 | -- | 18 | Group 21 | 30 - 60 | 010 |
| 03 0332-26 CM-8 | 033-57 | 0.985 | 0.065 | 0.103 | 0.075 | 2.35/2.40 | -- | 2.25/2.30 | 30 | -- | 18 | Group 21 | 25 - 60 | 000 |
| 0334-27 | 033-58 | 1.003 | 0.095 | 0.139 | 0.104 | 2.35/2.40 | -- | 2.35/2.40 | 30 | -- | 30 | Group 21 | 20 - 60 | 000 |
| 0334-28 CM-9 | 033-61 | 0.980 | 0.122 | 0.150 | 0.135 | $2.35^{+}$ | -- | 2.300/2.325 | 27 | -- | 22 | Group 10 | 20 - 60 | 000 |
| 0422-1 | 033-28 | 1.087 | 0.062 | 0.095 | 0.077 | $2.25^{+}$ | -- | 2.25/2.30 | 15 | -- | 18 | Group 21 (est.) | -- | -- |
| 0422-2 | 033-44 ATJ/A | 1.080 | 0.070 | 0.100 | 0.068 | -- | -- | 2.25/2.30 | -- | -- | 18 | Group 21 | -- | -- |
| 0532-1 | 033-13 | 1.106 | 0.070 | 0.085 | 0.073 | 2.35/2.40 | $2.40^{-}$ | $2.35^{+}$ | 30 | 32 | 27 | Group 10 | -- | -- |

[a] From known density/SiC relationship.

Table VI. 1.0-inch Throat Insert Data Summary
(fired nozzles only).

| Ident. No. | Deposition Run No. | Total Gas Flow Rate (SCFH) | Volume Percent $CH_4$ | Volume Percent MTS | Deposition Temperature (°F) | Deposition Rate (mils/hr) | | | Micro Code | SiC Percentage | |
|---|---|---|---|---|---|---|---|---|---|---|---|
| | | | | | | At Entrance | At Throat | At Exit | | At Entrance | At Exit |
| 0332-1 CM-1 | 162-25 | 149.8 | 1.34 | 0.21 | 3200 | 14.8 | 15.8 | 12.8 | -- | 30 | 24 |
| 0322-8 CM-2 | 162-54 | 149.8 | 1.34 | 0.21 | 3200 | 11.9 | 16.1 | 13.7 | 21 | 21 | 15 |
| 0332-10 (01) CM-3 | 162-66 | 149.8 | 1.34 | 0.21 | 3200 | 13.3 | 16.0 | 13.8 | 11/21 | 27 | 24 |
| 0332-12 CM-4 | 162-74 | 149.8 | 1.34 | 0.21 | 3200 | 11.4 | 15.5 | 14.7 | 21 | 27 | 27 |
| 0332-24 (02) CM-7 | 033-54 | 149.9 | 1.33 | 0.24 | 3200 | 10.7 | 16.6 | 13.0 | 20 | 30 | 18 |
| 0332-26 (03) CM-8 | 033-57 | 149.8 | 1.34 | 0.23 | 3200 | 10.5 | 16.6 | 12.1 | 21 | 30 | 18 |
| 0334-28 CM-9 | 033-61 | 149.8 | 1.34 | 0.21 | 3200 | 13.3 | 16.4 | 14.7 | 10 | 27 | 22 |
| AVERAGES | | 149.8 | 1.34 | 0.22 | 3200 | 12.3 | 16.2 | 13.7 | 21 | 27 | 21 |
| STD. DEV. | | | | | | 1.4 | 0.4 | 1.0 | -- | 3 | 4 |

Table VII. 1.0-Inch Insert Substrate Dimensional Data.

| Ident. No. | Deposition Run No. | Substrate Material | Percent Change in Substrate Dimensions After Coating | | | |
|---|---|---|---|---|---|---|
| | | | Entrance OD | Throat OD | Exit OD | Length |
| 0332-1 CM-1 | 162-25 | ATJ | 0 | -0.10 | 0 | +0.06 |
| 0332-2 | 162-27 | ATJ | -0.10 | -0.05 | 0 | -0.22 |
| 0322-4 | 162-43 | ATJ | -0.05 | 0 | -0.05 | -0.11 |
| 0332-5 | 162-45 | ATJ | -0.05 | -0.19 | 0 | 0 |
| 0322-6 | 162-48 | ATJ | 0 | 0 | -0.05 | -0.22 |
| 0332-7 | 162-52 | ATJ | +0.05 | 0 | +0.05 | -0.06 |
| 0322-8 CM-2 | 162-54 | ATJ | 0 | 0 | +0.05 | -0.11 |
| 01 0332-10 CM-3 | 162-66 | ATJ | 0 | +0.05 | +0.09 | -0.17 |
| 0322-11 | 162-73 | ATJ | 0 | +0.05 | 0 | +0.06 |
| 0332-12 CM-4 | 162-74 | ATJ/A | -0.14 | 0 | +0.05 | 0 |
| 0332-13 | 033-5 | PO-3 | -0.05 | -0.09 | 0 | -0.06 |
| 0332-14 | 033-8 | ATJS | +0.05[a] | +0.09[a] | +0.05[a] | +0.39[a] |

[a]Thin band of unalloyed PG at coating interface. Probably affected measurements.

Table VII. (continued).

| Ident. No. | Deposition Run No. | Substrate Material | Percent Change in Substrate Dimensions After Coating | | | |
|---|---|---|---|---|---|---|
| | | | Entrance OD | Throat OD | Exit OD | Length |
| 0322-15 | 033-21 | ATJ | ____[b] | ____[b] | ____[b] | ____[b] |
| 0322-16 | 033-22 | ATJ | ____[b] | ____[b] | ____[b] | ____[b] |
| 0322-17 | 033-23 | ATJ | ____[b] | ____[b] | ____[b] | ____[b] |
| 0322-18 | 033-24 | ATJS | ____[b] | ____[b] | ____[b] | ____[b] |
| 0322-20 | 033-27 | ATJ | ____[b] | ____[b] | ____[b] | ____[b] |
| 0312-21 | 033-29 | ATJ | ____[b] | ____[b] | ____[b] | ____[b] |
| 0312-22 | 033-33 | ATJ/A | 0 | +0.05 | +0.05 | 0 |
| 0336-23 | 033-50 | ATJ | +0.04 | -0.09 | 0 | -0.06 |
| 02 0332-24 CM-7 | 033-54 | ATJ | -0.09 | -0.09 | -0.19 | 0 |
| 0332-25 | 033-56 | ATJ | +0.05 | 0 | 0 | +0.06 |

[b] Dimensions were not measured with sufficient accuracy.

Table VII. (continued).

| Ident. No. | Deposition Run No. | Substrate Material | Percent Change in Substrate Dimensions After Coating | | | |
|---|---|---|---|---|---|---|
| | | | Entrance OD | Throat OD | Exit OD | Length |
| 03 0332-26 CM-8 | 033-57 | ATJ | -0.05 | -0.05 | -0.05 | -0.06 |
| 0334-27 | 033-58 | ATJ | -0.05 | -0.09 | -0.05 | +0.11 |
| 0334-28 CM-9 | 033-61 | ATJ | 0 | +0.05 | 0 | ____ |
| | | Average Change in ATJ | -0.02 | -0.03 | -0.01 | -0.05 |
| | | Standard deviation | 0.05 | 0.07 | 0.06 | 0.14 |

were taken of the entrance, throat and exit areas. Although good even SiC dispersion was obtained and good axial microstructural uniformity was achieved, the needles were shorter than expected. The third insert, 0332-3, was sectioned, circumferentially, at the entrance, throat and exit planes. These sections were then cut into four equal size quadrants and the density and SiC concentration of the coating determined for each. The results are given in Table VIII. These first three inserts, which were fabricated using an injector spacing of 3-1/4 inches, yielded coatings with an average coating thickness ratio of 0.89/1 between the entrance end and the throat. After the third run a new heater tube was installed in the furnace. The next two runs, 0322-4 and 0332-5, were conducted utilizing the same conditions as the first three runs. However, the entrance/throat coating thickness ratio dropped to 0.60/1. Apparently the installation of a new furnace tube affected the deposition rate profile in the deposition chamber. These changes are probably caused by changes in the longitudinal thermal gradient within the furnace tube which altered the thermal profile of the deposition chamber. To increase the entrance end coating thickness the injector spacing was increased from 3-1/4 to 3-3/4 inches. Other conditions remained the same. This increased the entrance/throat coating thickness ratio to an average of 0.75/1 through Run No. 0332-15, excluding No. 0322-11.

Run No. 0322-11 was conducted to determine if reducing the process nitrogen rate and increasing the deposition temperature would increase the SiC crystal size. The resultant coating was in the group 70 category indicating that these conditions were unsatisfactory.

The injector spacing was held at 3-3/4 during fabrication of all of the remaining standard throat inserts. An average entrance/throat coating thickness ratio of 0.75/1 was obtained on all of these inserts. However, considerable fluctuation in this ratio occurred throughout the program. Ratios as low as 0.63/1 and as high as 0.97/1 were encountered. No definite explanation of these fluctuations can be given. It is likely that they are related to longitudinal thermal gradients which always exist within the deposition chamber. These gradients do change with furnace use as changes in the heater tube thermal gradient occur. However, such changes usually take place over an extended period of time as the heater tube slowly degenerates and would not explain abrupt changes between runs. These are more likely caused whenever the deposition chamber is not returned to the same position in the furnace after each run. Since thermal gradient is obviously to exist within the furnace tube, any change in the location of the deposition chamber would result in changes of its temperature profile.

After 3-3/4 inches was established as the standard injector spacing a second insert, 0332-9, was sectioned in the same manner as 0332-3. These data are also shown in Table VIII.

Prior to Run No. 0322-15 bubbler weights were not recorded before and after deposition. After these weight recordings were begun it became apparent that the reproducibility of the $CH_3SiCl_3$ rate, from run to run, was less than desirable. This lack of reproducibility was found to be caused by leakage to the atmosphere, from the nitrogen inlet lines leading into the bubbler and between the bubbler and the injector. All of the lines and fittings from the nitrogen supply, through the bubbler, and into the injector were replaced. After Run No. 0322-18 these lines were leak tested prior to each deposition run.

At about this same time it was considered desirable to check the effect of temperature and gas velocity on the coating microstructure. To accomplish this purpose a deposition run, 03xx-19, was conducted in which the coating was deposited in five distinct layers, each separated by a thin PG band. The first four layers were deposited at 2,900, 3,000, 3,100 and 3,200°F, respectively. The fifth layer was deposited at 3,200°F with a 50 percent

Table VIII. 1.0-inch Throat Insert.

| Ident. No. | Deposition Run No. | Quadrant Measured (degrees) | Density (g/cc) | | | SiC Concentration (%) | | |
|---|---|---|---|---|---|---|---|---|
| | | | Entrance | Throat | Exit | Entrance | Throat | Exit |
| 0332-3 | 162-29 | 0 - 90 | 2.40$^{-}$ | 2.30/2.35 | 2.35$^{-}$ | 31[a] | 23[a] | 25[a] |
| | | 90 - 180 | 2.40 | 2.35/2.40 | 2.30$^{+}$ | 32[a] | 29[a] | 21[a] |
| | | 180 - 270 | 2.40 | 2.35/2.40 | 2.30/ 2.35 | 32[a] | 29[a] | 23[a] |
| | | 270 - 360 | 2.40 | 2.35/2.40 | 2.30/ 2.35 | 32[a] | 29[a] | 23[a] |
| 0332-9 | 162-58 | 0 - 90 | 2.35/ 2.40 | 2.35/2.40 | 2.35$^{-}$ | 20.0[b]<br>29[a] | 22.2[b]<br>29[a] | 18.9[b]<br>25[a] |
| | | 90 - 180 | 2.35/ 2.40 | 2.40$^{-}$ | 2.35/ 2.40 | 21.5[b]<br>29[a] | 23.2[b]<br>31[a] | 20.4[b]<br>29[a] |
| | | 180 - 270 | 2.35/ 2.40 | 2.35/2.40 | 2.35$^{-}$ | 20.7[b]<br>29[a] | 23.1[b]<br>29[a] | 21.0[b]<br>25[a] |
| | | 270 - 360 | 2.35$^{+}$ | 2.40$^{-}$ | 2.35/ 2.40 | 20.4[b]<br>27[a] | 23.8[b]<br>31[a] | 18.5[b]<br>29[a] |

[a] SiC concentration derived from density measurements.

[b] SiC concentration derived by ashing

increase in the process nitrogen flow rate to determine the effect of flow rate and turbulence on the coating microstructure. The results of this special deposition run are shown in Figure 8. The SiC particle size increases with increasing deposition temperature. The extreme coarseness of the fourth layer is not understood and more runs of this type should be made.

The next three runs, 0322-20, 0312-21 and 0312-22, were conducted at a reduced $CH_3SiCl_3$ rate to determine the compatibility of a PG/8% SiC coating with ATJ graphite. Only one of these runs, 0312-21, yielded a coating with a SiC concentration near that desired. The SiC phase was extremely fine and granular. No flaws were visible in either the coating or the substrate. Insert No. 0312-22 contained 12 percent SiC and was of similar microstructure. However, it contained a delamination within the coating near the substrate. This was believed to have been caused by variations in the $CH_3SiCl_3$ flow rate during deposition which, in turn, caused variations in the SiC concentration and induced excessive stresses within the coating.

The next deposition run, 0330-23, was conducted to fabricate a 200 mil thick coating. Post deposition examination showed the coating, near the substrate, to be of a coarse microstructure with a poor SiC dispersion. This band was 4 to 6 mils thick and contained a fine delamination. This situation had been encountered in the past and is believed to be caused by a lack of nucleation sites for the SiC during the initial phase of deposition. To provide these sites the $CH_3SiCl_3$ is introduced, into the furnace, 30 to 60 seconds before starting the $CH_4$ flow. Another cause of this type of anomaly is an insufficient soak time at deposition temperature before coating is started. At least 15 minutes of soak time are required to ensure that the deposition chamber has reached equilibrium before deposition is started.

The remaining five standard throat insert deposition runs, 0332-24 through 0334-28, were all conducted to fabricate inserts for motor test firings. Of these, three yielded coatings of acceptable quality.

Seven standard design 1.0-inch nozzle throat inserts were produced for test firing. Deposition conditions for these inserts were nearly identical as shown in Table VIII. The calculated linear flow rate of the reactant gases at the throat is 52 feet per second at the end of coating. Deposition rates were fairly consistent, around 15 mils per hour. The SiC content at the throat is higher than aimed (20 percent) because incomplete ashing during the SiC assay gave low answers. These SiC data are taken from the density/%SiC relationship and average 27 percent at the throat.

The wraparound design run results served to demonstrate that the fabrication of these coated shapes is feasible. No difficulties were encountered.

## 2. 1.0 INCH ENTRANCE AND EXIT SECTIONS

Eleven 1.0-inch entrance approach sections, Figure 9, and one 1.0-inch exit section, Figure 10, deposition runs were conducted. The first five, 0734-1 through 0712-5, were conducted using a 6-inch carbon tube furnace heated by a 100 kW, 10 Kc, Ajax motor generator. The remaining parts were fabricated in the 4-inch Pereny carbon tube resistance furnace. The deposition configuration for all twelve runs was similar and is shown in Figure 11.

1st Layer — 140 SCFH Process $N_2$.
2900°F deposition temperature.
Group 10-20 microstructure.
Dispersion is very good and SiC is acicular.

2nd Layer — 140 SCFH Process $N_2$.
3000°F deposition temperature.
Group 10-40 microstructure.
Crystals appear to have been cut across axis. This may explain the apparently small L/D ratio.
Note: Crystal length in lower right hand corner of picture.

3rd Layer — 140 SCFH Process $N_2$.
3100°F deposition temperature.
Group 11 microstructure.
Note: Poorer dispersion with SiC starting to concentrate in cone boundaries.

4th Layer — 140 SCFH Process $N_2$.
3200°F deposition temperature.
Group 30 microstructure.
SiC phase shows definite tendency to concentrate in cone boundaries.

5th Layer — 210 SCFH Process $N_2$.
3200° deposition temperature.
Group 30 microstructure.
Similar to 4th layer. Somewhat better dispersion may be the result of reduced deposition temperature which would be caused by cooling effect of increased $N_2$ flow rate.

Figure 8. 1.0-Inch Throat Insert Fabricated Using Variable Deposition Conditions. All Views are from Exit End of 03XX-19 at 800X.

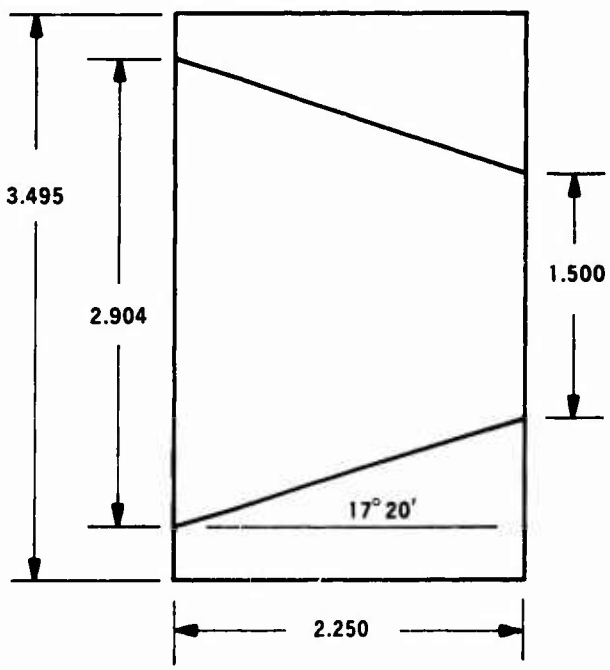



Figure 9. 1.0-Inch Entrance Approach Section Substrate.

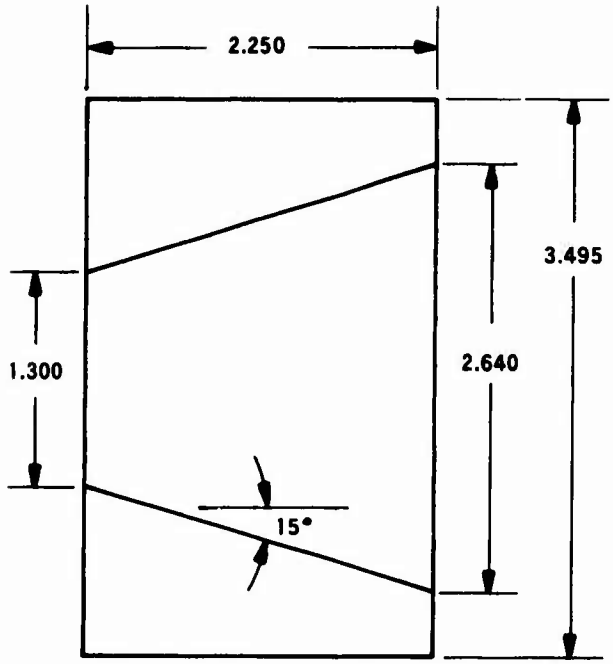



Figure 10. 1.0-Inch Exit Section Substrate.

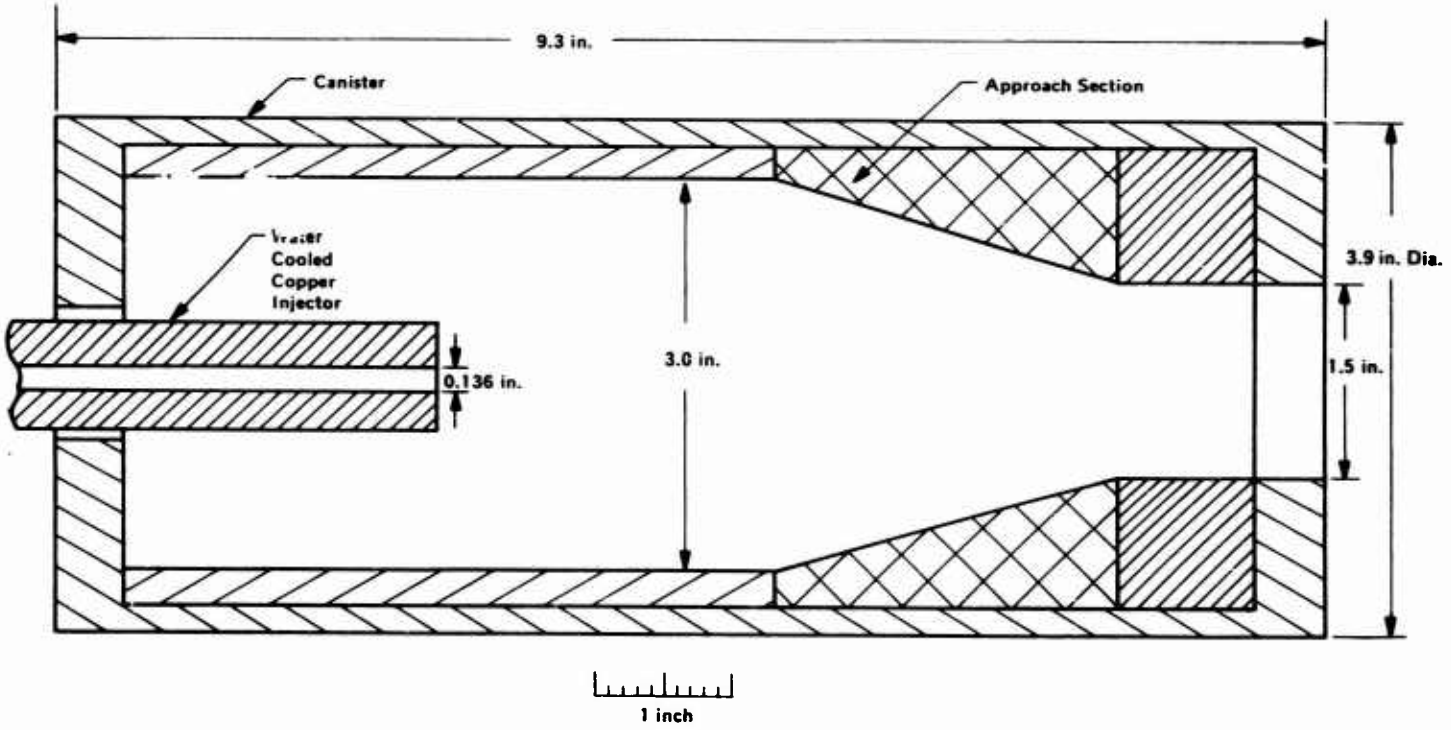



Figure 11. 1.0-inch Entrance Approach and Exit Section Deposition Assembly.

Coating process conditions are shown in Table IX and coating characterization data are shown in Table X.

Additional process and coating data for the seven parts that were fired are summarized in Table XI.

All substrates were fabricated from ATJ graphite with the throat axis normal to the grain direction.

Substrate dimensions were recorded before and after coating. The data are shown in Table XII. There appeared to be a slight decrease in the average substrate outside diameter and a slight increase in the length after coating. All permanent substrate outside diameter deformations recorded were between -0.1 and +0.04 percent. However, the data are fragmentary, and because of this, it is difficult to reach any definite conclusions.

In the first five runs, which were conducted in the 6-inch induction furnace, the SiC concentration, at the exit end, decreased steadily from a high of 30 percent in the first run, 0734-1, to a low of 12 percent in the fifth run, 0712-5. This was undoubtedly related to the continuous degeneration of the furnace tube as described in Section VII. After the loss of the 6-inch furnace tube the process was changed to the 4-inch Pereny furnace. At this time the deposition temperature was reduced from 3,350°F to 3,200°F and the total process gas flow rate was increased by 22 percent by increasing the process nitrogen flow rate. The remaining seven runs were conducted with no serious problems.

One exit section, 0934-1, was fabricated using the same parameters as the latter entrance approach sections.

It may be noted that the coating rate decreased and the SiC content at the exit end increased when the switch was made to the Pereny furnace. The rate decrease and part of the SiC increase may be attributed to the decrease in $CH_4$ content of the reactant gases. The main cause of the increase in SiC content is probably due to the decrease in temperature. On the other hand, the SiC needles coarsened when the process was switched to the Pereny furnace. This cannot be explained at this time. However, the latter Pereny run microstructures are consistent with the 1.0-inch throat insert microstructures.

### 3. 1.72-INCH HIPPO NOZZLE THROAT INSERTS

Forty-four standard (see Figure 12) and three wraparound (see Figure 13) 1.72-inch HIPPO nozzle throat insert deposition runs were conducted. The deposition assembly configuration is shown in Figure 14. All except the first four and the last two runs utilized a Pereny 4-inch carbon tube resistance furnace as the heat source.

Coating process conditions are shown in Table XIII and coating characterization data are shown in Table XIV. Additional process and coating data for the eight fired throat inserts are summarized in Table XV.

The coating microstructure of all the 1.72-inch inserts, fabricated in the 4-inch Pereny furnace, were similar and were mostly code 20.

Most of the substrates were fabricated of ATJ graphite and machined with the axis normal to the grain direction. A few HLM substrates were coated for sectioning as reproducibility specimens. Two annealed ATJ substrates were also coated. The substrate material used, if other than ATJ, is listed below the deposition run number in Table XIII.

## Table IX. 1.0-inch Entrance Approach and Exit Section Coating Process Conditions.

| Ident. No. | Deposition Run No. | Annulus $N_2$ (SCFH) | Process $N_2$ (SCFH) | Process $CH_4$ (SCFH) | Process $N_2$ to Bubbler (SCFH) | $CH_3SiCl_3$ Rate (g/hr) | Deposition Temperature (°F) | Deposition Time (hr) | Injector Spacing (in.) | Remarks |
|---|---|---|---|---|---|---|---|---|---|---|
| 0734-1 | 001-21 | 5.0 | 114 | 4.0 | 5.0 | 108[a] | 3,350 | 5.50 | 5 | 6-inch induction furnace. |
| 0722-2 CM-3 | 001-23 | 5.0 | 114 | 4.0 | 5.0 | 108[a] | 3,350 | 4.17 | 5 | Conducted to fabricate entrance approach section for test firing. |
| 0722-3 CM-4 | 001-27 | 5.0 | 114 | 4.0 | 5.0 | 108[a] | 3,350 | 4.00 | 5 | Same as above. |
| 0722-4 CM-5 | 001-30 | 5.0 | 114 | 4.0 | 5.0 | 108[a] | 3,350 | 4.00 | 5 | Same as above. |
| 0712-5 | 001-33 | 5.0 | 114 | 4.0 | 5.0 | 108[a] | 3,350 | 4.00 | 5 | Temperature readings may have been in error. Furnace tube going bad. |
| 0734-6 | 033-46 | 5.0 | 150 | 4.0 | 5.0 | 118 | 3,200 | 8.00 | 5 | Changed to 4-inch Peremy furnace. Process $N_2$ rate raised and deposition temperature lowered to increase SiC dispersion. |
| 02 0732-7 CM-7 | 033-52 | 5.0 | 150 | 4.0 | 5.0 | 125 | 3,200 | 5.00 | 5 | Deposition time reduced to reduce coating thickness. |
| 0732-8 | 033-53 | 5.0 | 150 | 4.0 | 5.0 | 127 | 3,200 | 3.75 | 5 | Deposition time reduced to reduce coating thickness. |
| 0732-9 | 033-55 | 5.0 | 150 | 4.0 | 5.0 | 117 | 3,200 | 4.50 | 5 | MTS rate reduced inadvertently during early part of cycle. |
| 0734-10 CM-9 | 033-60 | 5.0 | 150 | 4.0 | 5.0 | 120 | 3,200 | 7.00 | 5 | Increased deposition time to achieve 150 mil coating thickness. |
| 03 0722-11 CM-8 | 033-73 | 5.0 | 150 | 4.0 | 4.6 | 96 | 3,200 | 5.33 | 5 | $N_2$ to bubbler rate reduced to reduce SiC concentration. Deposition time reduced to achieve 100 mil coating thickness. |
| 03 0934-1 CM-8 | 033-59 | 5.0 | 150 | 4.0 | 5.0 | 98 | 3,200 | 5.50 | 5 | Exit section. Similar to entrance approach sections but diameters reduced to match exit end of nozzle insert. |

[a]Calculated assuming saturation of bubbler $N_2$ at 90°F.

## Table X. 1.0-inch Entrance Approach and Exit Section Coating Evaluation Data.

| Ident No. | Deposition Run No. | Coating Thickness (in.) | | Density (gm/cc) | | SiC[a] Concentration (%) | | Appearance | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | | | | | | | | Entrance | | | Exit | | |
| | | Entrance | Exit | Entrance | Exit | Entrance | Exit | Micro-Structure | Cone Angle (degree) | Surface Condition | Micro-Structure | Cone Angle (degree) | Surface Condition |
| 0734-1 | 001-21 | 0.090 | 0.132 | 2.30 | 2.35/2.40 | 21 | 30 | Contains band of low SiC concentration | | | Group 11 (est.) No visible flaws. | | |
| 0722-2 CM-3 | 001-23 | 0.099 | 0.118 | 2.25 | 2.30 | 15 | 21 | Group 30 | | | Group 10 (est.) | | |
| 0722-3 CM-4 | 001-27 | 0.048 | 0.113 | 2.25 | 2.25 | 15 | 15 | Group 60 | | | Group 11 | | |
| 0722-4 CM-5 | 001-30 | 0.025 | 0.125 | — | 2.25 | — | 15 | Group 60 (est.) | | | Group 11 (est.) | | |
| 0712-5 | 001-33 | 0.040 | 0.113 | 1.95 | 2.20/2.25 | 0 | 12 | Group 60 | | | Group 60 | | |
| 0734-6 | 033-46 | — | 0.143 | — | 2.35/2.40 | — | 30 | Not examined | | | Fine discontinuous delamination visible near interface. Group 21 | | |
| 02 0732-7 CM-7 | 033-52 | 0.038 | 0.107 | — | 2.40/2.45 | — | 34 | Group 60 | | | Group 21 | | |
| 0732-8 | 033-53 | 0.047 | 0.097 | 2.30/2.35 | 2.35/2.40 | 24 | 30 | Group 70 | | | Group 21 Coating contains occasional intraconical delaminations. | | |
| 0732-9 | 033-55 | 0.036 | 0.107 | — | 2.35/2.40 | — | 30 | Group 60 | 35 - 75 | 110 | Group 21 | 30 - 50 | 110 |
| 0734-10 CM-9 | 033-60 | 0.066 | 0.145 | 2.30 | 2.40/2.45 | 21 | 34 | Unalloyed PG layer visible at interface at both ends. Group 60 | 20 - 40 | 200 | Group 20 | 30 - 65 | 200 |
| 03 0722-11 CM-8 | 033-73 | 0.038/0.052 | 0.113 | 2.25/2.30 | 2.275/ 2.300 | 18 | 19 | Group 70 | 40 - 80 | 110 | Group 21 | 40 - 60 | 110 |
| 03 0934-1 CM-8 | 033-59 | 0.088 | 0.141 | 2.300/ 2.325 | 2.35/2.40 | 22 | 30 | Group 60 | 20 - 40 | 300 | Group 20 | 30 - 55 | 300 |

[a] Determined by density/SiC content relationship.

Table XI. 1.0-inch Entrance Approach and Exit Section Data Summary (fired nozzles only).

| Ident. No. | Deposition Run No. | Total Gas Flow Rate (SCFH) | Volume Percent $CH_4$ | Volume Percent MTS | Deposition Temperature (°F) | Deposition Rate (mils/hr) | | Micro Code | SiC Percentage | |
|---|---|---|---|---|---|---|---|---|---|---|
| | | | | | | At Entrance | At Exit | | At Entrance | At Exit |
| 0722-2(01) CM-3 | 001-23 | 129 | 3.11 | 0.48 | 3350 | 23.7 | 28.3 | 10 | 15 | 21 |
| 0722-3 CM-4 | 001-27 | 129 | 3.11 | 0.48 | 3350 | 12.0 | 28.3 | 11 | 15 | 15 |
| 0722-4 CM-5 | 001-30 | 129 | 3.11 | 0.48 | 3350 | 6.2 | 31.3 | 11 | -- | 15 |
| | SWITCH TO PERENY FURNACE | | | | | | | | | |
| 0732-7(02) CM-7[a] | 033-52 | 165 | 2.43 | 0.43 | 3200 | 7.6 | 21.4 | 21 | -- | 34 |
| 0734-10 CM-9 | 033-60 | 165 | 2.43 | 0.42 | 3200 | 9.4 | 20.7 | 20 | 21 | 34 |
| 0722-11(03) CM-8 | 033-73 | 164 | 2.44 | 0.33 | 3200 | 8.4 | 21.2 | 21 | 18 | 19 |
| 0934-1(03) CM-8 Exit Section | 033-59 | 165 | 2.43 | 0.34 | 3200 | 16.0 | 25.6 | 20 | 22 | 30 |

[a] CM-6 was a graded coating and is described in Phase I - Task 2.

Table XII. 1.0-Inch Entr. ce Approach and Exit Section Substrate Dimensional Data.

| Ident. No. | Deposition Run No. | Substrate Material | Percent Change in Substrate Dimensions After Coating | | | |
|---|---|---|---|---|---|---|
| | | | Entrance OD | Mid OD | Exit OD | Length |
| 0734-6 | 033-46 | ATJ | 0 | 0 | 0 | 0 |
| 02<br>0732-7<br>CM-7 | 033-52 | ATJ | 0 | +0.03 | +0.03 | +0.04 |
| 0732-8 | 033-53 | ATJ | +0.03 | 0 | +0.03 | +0.04 |
| 0732-9 | 033-55 | ATJ | 0 | +0.03 | 0 | +0.05 |
| 0734-10<br>CM-9 | 033-60 | ATJ | -0.14 | +0.03 | -0.14 | +0.22 |
| 03<br>0934-1<br>CM-8 | 033-59 | ATJ | +0.03 | -0.03 | 0 | -0.09 |
| | Average Dimensional Changes | | -0.01 | +0.01 | -0.01 | +0.04 |

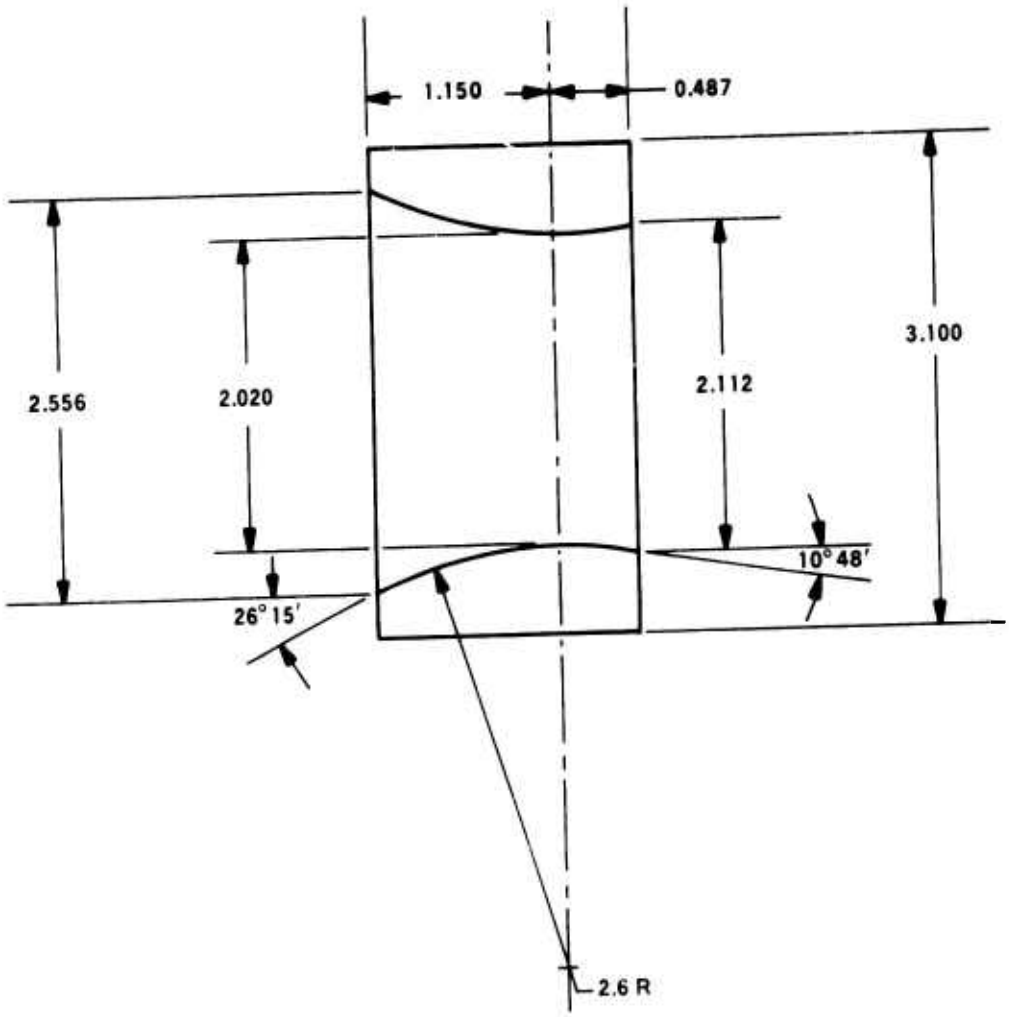



Figure 12. 1.72-Inch Standard Nozzle Throat Insert Substrate.

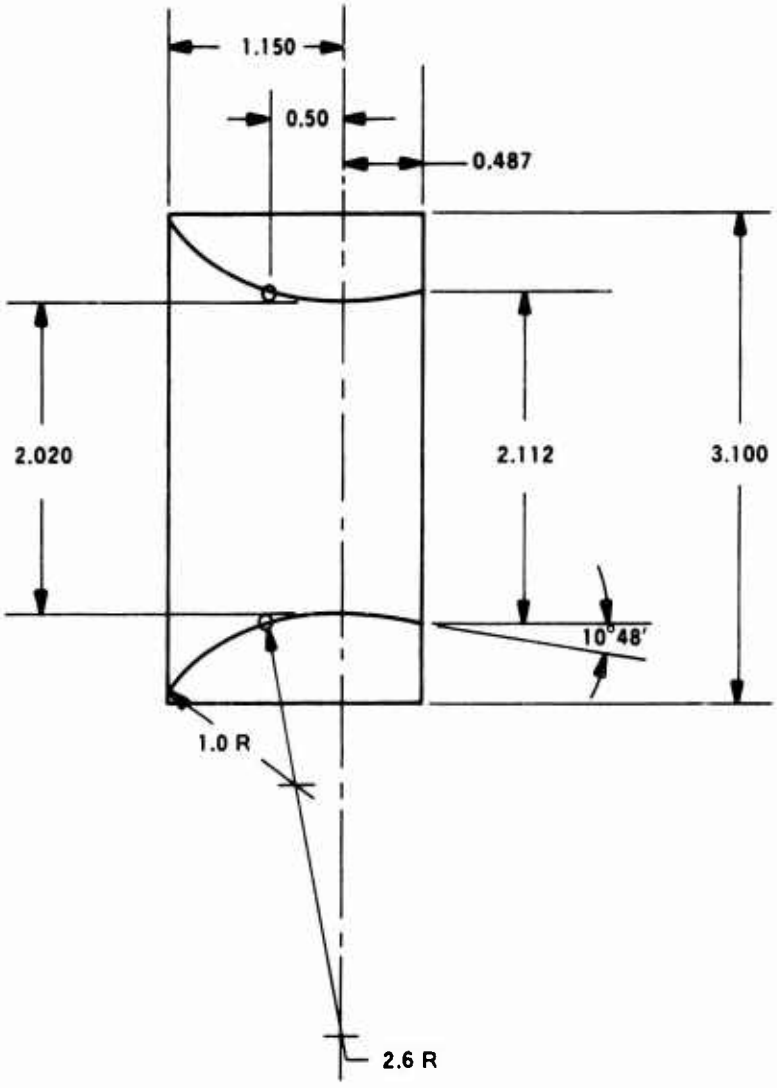



Figure 13. 1.72-Inch Wraparound Nozzle Throat Insert Substrate.

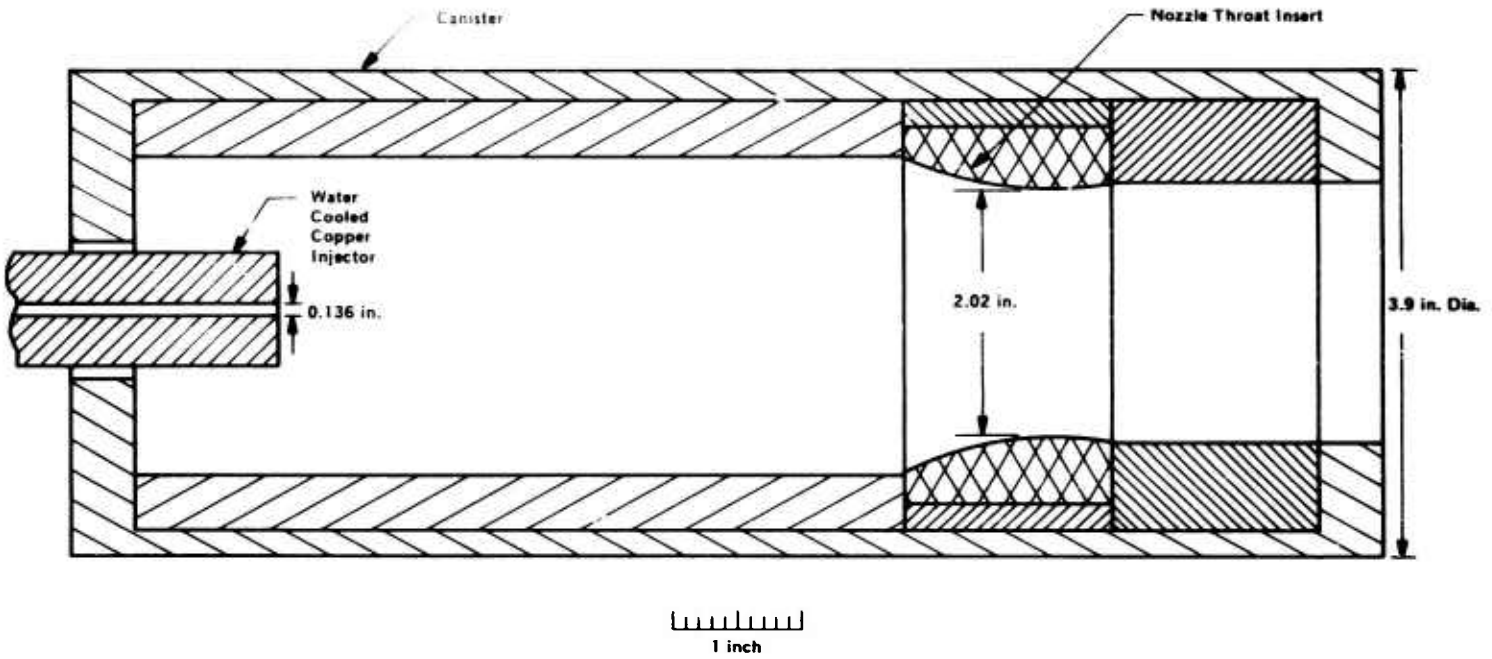



**Figure 14. 1.72-inch HIPPO Throat Insert Deposition Assembly.**

## Table XIII. 1.72-inch Throat Insert Coating Process Conditions.

| Ident. No. | Deposition Run No. | Annulus $N_2$ (SCFH) | Process $N_2$ (SCFH) | Process $CH_4$ (SCFH) | Process $N_2$ to Bubbler (SCFH) | $CH_3SiCl_3$ Rate (g/hr) | Deposition Temperature (°F) | Deposition Time (hr) | Injector Spacing (in.) | Remarks |
|---|---|---|---|---|---|---|---|---|---|---|
| 23XX-1 | 001-24 | 5.0 | 114 | 4.0 | 5.0 | - | 3,350 | 4.0 | - | First 1.720-inch coating run - in 6 inch induction furnace |
| 2322-2 | 001-25 | 5.0 | 180 | 4.0 | 5.0 | - | 3,350 | 4.0 | - | PG precoated substrate so coating could be removed and examined |
| 23XX-3 | 001-29 | Disregard. Wrong material used for MTS. | | | | | | | | |
| 2312-4 | 001-32 | 5.0 | 114 | 4.0 | 5.0 | - | 3,350 | 4.0 | - | Wrong process $N_2$ rate used. Was to have been 180 SCFH |
| 2332-5 | 033-18 | 5.0 | 150 | 4.0 | 5.0 | - | 3,200 | 5.0 | 5 3/8 | PG precoated substrate. Deposition temperature reduced to improve SiC dispersion. DOW MTS used as in past. Switched to 4-inch Peremy furnace |
| 2322-6 | 033-20 | 5.0 | 150 | 4.0 | 5.0 | 105 | 3,200 | 5.0 | 5 3/8 | Same as 033-18 utilizing Union Carbide MTS |
| 2322-7 | 033-38 ATJ/A | 5.0 | 150 | 4.0 | 4.6 | - | 3,200 | 5.0 | 5 | Bubbler $N_2$ rate reduced to lower SiC concentration to 15%. Injector moved forward to improve SiC dispersion. |
| 2322-8 | 033-39 ATJ/A | 5.0 | 150 | 4.0 | 4.6 | 99 | 3,200 | 3.5 | 5 | Deposition time reduced to reduce coating thickness to 100 mils |
| 2322-9 (0001) | 033-40 | 5.0 | 150 | 4.0 | 4.6 | 110 | 3,200 | 4.50 | 5 | Deposition time increased to increase coating thickness to 100 mils |
| 2322-10 | 033-41 | 5.0 | 150 | 4.0 | 4.6 | 100 | 3,200 | 4.50 | 5 | None |
| 2322-11 | 033-45 | 5.0 | 150 | 4.0 | 4.6 | 110 | 3,200 | 4.50 | 4 | Tapered substrate approach section used. Injector inadvertently placed 1.0 inch too close to throat. |
| 2332-12 | 033-47 | 5.0 | 150 | 4.0 | 5.0 | 121 | 3,200 | 4.37 | 5 1/4 | Injector moved back 1 1/4 inch to increase coating thickness at entrance end. |
| 2332-13 | 033-48 | 5.0 | 150 | 4.0 | 5.0 | 114 | 3,200 | 4.83 | 5 7/8 | Injector spacing increased to increase coating thickness at entr. end. |
| 2322-14 | 033-49 | 5.0 | 150 | 4.0 | 5.0 | 115 | 3,200 | 4.50 | 5 3/8 | Returned to parameters of 033-18 to check controls. |
| 2332-15 | 033-51 | 5.0 | 150 | 4.0 | 5.0 | 110 | 3,200 | 4.97 | 5 3/8 | Cleaned injector and all associated gas introduction hardware and repeat previous run. |

## Table XIII. (continued)

| Ident. No. | Deposition Run No. | Annulus $N_2$ (SCFH) | Process $N_2$ (SCFH) | Process $CH_4$ (SCFH) | Process $N_2$ to Bubbler (SCFH) | $CH_3SiCl_3$ Rate (g/hr) | Deposition Temperature (°F) | Deposition Time (hr) | Injector Spacing (in.) | Remarks |
|---|---|---|---|---|---|---|---|---|---|---|
| 2322-16 | 033-62 | 5.0 | 150 | 4.0 | 5.0 | 102.6 | 3,200 | 4.67 | 5 3/8 | None |
| 2332-17 | 033-63 | 5.0 | 150 | 4.0 | 5.0 | 113.4 | 3,200 | 4.68 | 5 3/8 | None |
| 2322-18 (0002) | 033-65 | 5.0 | 150 | 4.0 | 5.0 | 96.8 | 3,200 | 4.75 | 5 3/8 | None |
| 2322-19 | 319-1 | 5.0 | 150 | 4.0 | 5.0 | 130.6 | 3,200 | 4.67 | 5 3/8 | None |
| 2322-20 | 319-8 | 5.0 | 150 | 4.0 | 5.0 | 87.3 | 3,200 | 4.67 | 5 3/8 | None |
| 2322-21 | 319-9 | 5.0 | 150 | 4.0 | 5.0 | 97.7 | 3,200 | 4.58 | 5 | Injector inadvertently placed at 5 inches instead of 5 3/8 as planned. |
| 2322-22 | 319.10 | 5.0 | 150 | 4.0 | 5.0 | 110.2 | 3,200 | 4.70 | 5 3/8 | None |
| 2332-23 (0003) | 319-11 | 5.0 | 150 | 4.0 | 5.0 | 103.0 | 3,200 | 4.75 | 5 3/8 | None |
| 2322-24 | 319-12 | 5.0 | 150 | 4.0 | 5.0 | 108.4 | 3,200 | 4.75 | 5 3/8 | None |
| 2334-25 | 319-17 | 5.0 | 150 | 4.0 | 5.0 | 128.7 | 3,200 | 7.25 | 5 3/8 | None |
| 2324-26 | 319-19 | 5.0 | 150 | 4.0 | 5.0 | 134.9 | 3,200 | 7.25 | 6 | Injector spacing increased to 6 inches to increase coating thickness at entrance end. |
| 2334-27 (0004) | 319-20 | 5.0 | 230 | 4.0 | 5.0 | 125.6 | 3,200 | 7.25 | 6 | Process $N_2$ rate increased to improve coating surface condition. |
| 2334-28 | 319-24 | 5.0 | 210 | 4.0 | 4.6 | 124.6 | 3,200 | 7.5 | 6 | MTS rate reduced to reduce SiC concentration. Process $N_2$ rate reduced because flowmeter cannot control at 230 SCFH. |
| 2324-29 | 319-33 | 5.0 | 210 | 4.0 | 4.0 | 123.8 | 3,200 | 7.75 | 6 | MTS rate reduced to reduce SiC concentration. |
| 2324-30 (Repr) | 319-41 HLM | 5.0 | 210 | 4.0 | 4.0 | 91 (approx.) | 3,200 | 7.75 | 6 | PG precoated substrate - coating was used for reproducibility studies |
| 2324-31 | 319-42 | 5.0 | 210 | 4.0 | 4.0 | 105.7 | 3,200 | 7.92 | 6 | Exhaust tube plugged near end of run. |
| 2324-32 | 319-43 | 5.0 | 210 | 4.0 | 4.0 | 100.7 | 3,200 | 7.50 | 6 | Deposition time reduced to reduce coating thickness. |
| 2324-33 | 319-44 | 5.0 | 210 | 4.0 | 4.0 | 101.3 | 3,200 | 7.85 | 6 | Exhaust tube plugged near end of run |

## Table XIII. (continued)

| Ident. No. | Deposition Run No. | Annulus $N_2$ (SCFH) | Process $N_2$ (SCFH) | Process $CH_4$ (SCFH) | Process $N_2$ to Bubbler (SCFH) | $CH_3SiCl_3$ Rate (g/hr) | Deposition Temperature (°F) | Deposition Time (hr) | Injector Spacing (in.) | Remarks |
|---|---|---|---|---|---|---|---|---|---|---|
| 2324-34 | 319-45 HLM | 5.0 | 230 | 4.0 | 4.0 | 109.2 | 3,200 | 7.40 | 6 | New process $N_2$ flowmeter installed so higher rates can be used to improve coating surface condition. |
| 2324-35 (0005) | 319-46 | 5.0 | 230 | 4.0 | 4.0 | 103.3 | 3,200 | 7.65 | 6 | None |
| 2324-36 | 319-48 HLM | 5.0 | 230 | 4.0 | 4.0 | - | 3,200 | - | 6 | PG precoated substrate. Run turned off at wrong time. Deposition time unknown. |
| 2324-37 (0006) | 319-50 | 5.0 | 230 | 4.0 | 4.0 | 109.2 | 3,200 | 7.65 | 6 | None |
| 2324-38 (Repr) | 319-53 HLM | 5.0 | 230 | 4.0 | 4.0 | 102.2 | 3,200 | 7.65 | 6 | PG precoated substrate. |
| 2324-39 (Repr) | 319-54 HLM | 5.0 | 230 | 4.0 | 4.0 | 91.6 | 3,200 | 7.65 | 6 | PG precoated substrate. coating was used for reproducibility studies. |
| 2324-40 | 002-2 | 5.0 | 230 | 4.0 | 4.0 | 104.4 | 3,200 | 6.25 | 6 | Run terminated early. Sight tube became blocked causing overheating. |
| 2334-41 | 002-4 | 5.0 | 230 | 4.0 | 4.0 | 97.4 | 3,200 | 8.45 | 6 | New $N_2$ bubbler flowmeter installed. MTS bubbler overfilled. |
| 2334-42 (0007) | 002-7 | 5.0 | 230 | 4.0 | 4.0 | 94.1 | 3,200 | 8.45 | 6 | Reduced liquid level in bubbler. |
| 2322-43 | 002-10 | 5.0 | 230 | 4.0 | 4.0 | 97.3 | 3,200 | 8.45 | 6 | Run conducted in 6-inch Feremy because 4-inch Feremy was inoperable. |
| 2324-44 (0008) | 002-12 | 5.0 | 230 | 4.0 | 4.0 | 97.3 | 3,200 | 8.45 | 6 1/2 | Injector moved back to increase coating thickness at entrance end and throat. 6-inch Feremy used again. |
| 2424-1 | 319-47 HLM | 5.0 | 230 | 4.0 | 4.0 | 100.8 | 3,200 | 7.65 | 6 | PG precoated substrate. |
| 2422-2 (Repr) | 319-49 HLM | 5.0 | 230 | 4.0 | 4.0 | 99.2 | 3,200 | 5.27 | 6 3/4 | PG precoated substrate. Injector moved back to increase coating thickness at entrance end. |
| 2424-3 (Repr) | 319-51 HLM | 5.0 | 230 | 4.0 | 4.0 | 102.4 | 3,200 | 7.65 | 6 3/4 | PG precoated substrate. |

## Table XIV. 1.72-inch HIPPO Throat Insert Coating Evaluation Data.

| Ident. No. | Deposition Run No. | Coated Throat Diameter (in.) | Coating Thickness (in.) | | | Density (gm/cc) | | | SiC Concentration (%) | | | Appearance | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | | | Entrance | Throat | Exit | Entrance | Throat | Exit | Entrance | Throat | Exit | Microstructure | Cone Angle (degree) | Surface Condition |
| 23XX-1 | 001-24 | — | — | — | — | — | — | — | — | — | — | Coating surface very rough and covered with clinker-like growths. No useful measurements possible. | | |
| 2322-2 | 001-25 | 1.717 | 0.067 | 0.089 | 0.083 | 2.25+ | 2.25/2.30 | 2.25+ | 15 | 18 | 15 | Group 11 (est.) | | |
| 23XX-3 | 001-29 | — | No SiC present. Wrong material used in place of MTS. | | | | | | | | | | | |
| 2312-4 | 001-32 | 1.760 | 0.059 | 0.080 | 0.069 | 2.20/2.25 | — | 2.15/2.20 | 12 | — | 3 | | | |
| 2332-5 | 033-18 | 1.690 | 0.085 | 0.118 | 0.094 | 2.35 | 2.30 | 2.30 | 27 | 21 | 21 | | | |
| 2322-6 | 033-20 | 1.697 | 0.098 | 0.111 | 0.089 | 2.30 | 2.30 | 2.25/2.30 | 21 | 21 | 18 | | | |
| 2322-7 | 033-38 | 1.664 | 0.060 | 0.118 | 0.108 | — | — | — | — | — | — | | | |
| 2322-8 | 033-39 | 1.761 | 0.074 | 0.080 | 0.072 | 2.25/2.30 | — | 2.25/2.30 | 18 | — | 18 | | | |
| 2322-9 (0001) | 033-40 | 1.716 | 0.092 | 0.102 | 0.094 | 2.25/2.30 | — | 2.30/2.35 | 18 | — | 24 | Group 21 | | |
| 2322-10 | 033-41 | 1.732 | 0.053 | 0.094 | 0.084 | 2.20/2.25 | — | 2.30/2.35 | 12 | — | 24 | Group 60 - Ent.<br>Group 21 - Exit | | |
| | | | | | | | | | | | | No SiC visible in first 30% of coating | | |
| 2322-11 | 033-45 | 1.745 | 0.027 | 0.087 | 0.089 | 2.20/2.25 | — | 2.35/2.40 | 12 | — | 30 | Group 60 - Ent.<br>Group 21 - Exit | | Rough |
| 2332-12 | 033-47 | 1.738 | 0.047 | 0.091 | 0.078 | 2.30/2.35 | — | 2.35/2.40 | 24 | — | 30 | Group 30 - Ent.<br>Group 21 - Exit | | Rough |
| 2332-13 | 033-48 | 1.709 | 0.073 | 0.106 | 0.094 | 2.35/2.40 | — | 2.35/2.40 | 30 | — | 30 | Group 30 - Ent.<br>Group 21 - Exit | | Rough |
| 2322-14 | 033-49 | 1.729 | 0.058 | 0.095 | 0.077 | 2.25/2.30 | — | 2.30/2.35 | 18 | — | 24 | Group 30 - Ent.<br>Group 21 - Exit | | Rough |
| 2332-15 | 033-51 | 1.701 | 0.075 | 0.108 | 0.094 | 2.25/2.30 | — | 2.35/2.40 | 18 | — | 30 | Group 21 - Ent. | | Rough |
| 2322-16 | 033-62 | 1.720 | 0.071 | 0.100 | 0.087 | 2.275 | — | 2.275/2.300 | 18 | — | 19 | Group 20 | 35 - 50 | 110 |
| 2332-17 | 033-63 | 1.709 | 0.089 | 0.106 | 0.094 | 2.300/2.325 | — | 2.300/2.325 | 23 | — | 23 | Group 20 | 35 - 65 | Rough |
| 2322-18 (0002) | 033-65 | 1.719 | 0.077 | 0.098 | 0.094 | 2.300 | — | 2.275/2.300 | 21 | — | 19 | Group 20 | | 122 |
| 2322-19 | 319-1 | 1.730 | 0.051 | 0.095 | 0.079 | 2.250/2.275 | — | 2.300 | 16 | — | 21 | Group 30 | 40 - 60 | 100 |
| 2322-20 | 319-8 | 1.710 | 0.084 | 0.105 | 0.095 | 2.275 | — | 2.250/2.275 | 18 | — | 16 | Group 30 | 40 - 80 (60 avg.) | 110/300 |
| 2322-21 | 319-9 | 1.724 | 0.074 | 0.098 | 0.089 | 2.250/2.275 | — | 2.250 | 16 | — | 15 | Group 30 | 45 - 55 | 210 |
| 2322-22 | 319-10 | 1.740 | 0.037 | 0.090 | 0.089 | 2.275 | — | 2.30 | 18 | — | 21 | Group 20 - Ent.<br>Group 30 - Exit | 45 - 60 | 111 |
| 2332-23 (0003) | 319-11 | 1.720 | 0.077 | 0.100 | 0.091 | 2.300 | — | 2.300/2.325 | 21 | — | 23 | Group 30 - Ent.<br>Group 20 - Exit | 45 - 65 (55 avg.) | 210 |
| 2322-24 | 319-12 | 1.720 | 0.070 | 0.100 | 0.091 | 2.275/2.300 | — | 2.275/2.300 | 19 | — | 19 | Group 30 - Ent.<br>Group 20 - Exit | 40 - 65 (50 avg.) | 110 |
| 2334-25 | 319-17 | 1.730 | 0.097 | 0.143 | 0.143 | 2.300 | — | 2.300/2.325 | 21 | — | 23 | Group 30 | 40 - 55 | 211 |
| 2324-26 | 319-19 | 1.705 | 0.138 | 0.158 | 0.143 | 2.275/2.300 | — | 2.300 | 19 | — | 21 | Group 30 | 45 - 65 (55 avg.) | 222 |

## Table XIV. (continued)

| Ident. No. | Deposition Run No. | Coated Throat Diameter (in.) | Coating Thickness (in.) | | | Density (gm/cc) | | | SiC Concentration (%) | | | Appearance | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | | | Entrance | Throat | Exit | Entrance | Throat | Exit | Entrance | Throat | Exit | Microstructure | Cone Angle (degree) | Surface Condition |
| 2334-27 (0004) | 319-20 | 1.721 | 0.138 | 0.150 | 0.139 | 2.325 | — | 2.325 | 24 | — | 24 | Group 20 | 30 - 50 (40 avg.) | 200 |
| 2334-28 | 319-24 | 1.713 | 0.135 | 0.154 | 0.146 | 2.30/2.35 | — | 2.35/2.40 | 24 | — | 30 | Group 21 - Ent.<br>Group 20 - Exit | 60 | 010 |
| 2324-29 | 319-33 | 1.728 | 0.129 | 0.146 | 0.137 | 2.30 | — | 2.30 | 21 | — | 21 | Group 21 - Ent.<br>Group 20 - Exit | 50 - 70<br>40 - 70 (exit) | 300 |
| 2324-30 (Repr.) | 319-41 | 1.72 | 0.159 | 0.165 | 0.152 | 2.25/2.30 | — | 2.25/2.30 | 18 | — | 18 | Group 21 - Ent.<br>Group 20 - Exit | 75 (ent.)<br>55 (exit) | 310 |
| 2324-31 | 319-42 | 1.700 | 0.142 | 0.160 | 0.143 | 2.25/2.30 | — | 2.25/2.30 | 18 | — | 18 | Group 20 | 50 | 300 |
| 2324-32 | 319-43 | 1.734 | 0.107 | 0.143 | 0.129 | 2.25/2.30 | — | 2.25/2.30 | 18 | — | 18 | Group 30 - Ent.<br>Group 20 - Exit | 60 - 70 (ent.)<br>45 - 60 (exit) | 300 |
| 2324-33 | 319-44 | 1.694 | 0.129 | 0.163 | 0.148 | 2.25/2.30 | — | 2.25/2.30 | 18 | — | 18 | Group 21 | 55 (ent.)<br>40 (exit) | 300 |
| 2324-34 | 319-45 | 1.630 | 0.125 | 0.145 | 0.134 | 2.25/2.30 | — | 2.30* | 18 | — | 22 | Group 21 - Ent.<br>Group 20 - Exit | 60 (ent.)<br>40 (exit) | 310 |
| 2324-35 (0005) | 319-46 | 1.715 | 0.135 | 0.152 | 0.142 | 2.25/2.30 | — | 2.25/2.30 | 18 | — | 18 | Group 20 | 60 (ent.)<br>40 (exit) | 300 |
| 2324-36 | 319-48 | 1.580 | 0.166 | 0.170 | 0.159 | 2.25/2.30 | — | 2.25/2.30 | 18 | — | 18 | Group 20 | 50 (ent.)<br>40 (exit) | 200 |
| 2324-37 (0006) | 319-50 | 1.724 | 0.142 | 0.148 | 0.139 | 2.25/2.30 | — | 2.30 | 18 | — | 21 | Group 20 | 75 (ent.)<br>45 (exit) | 110 |
| 2324-38 | 319-53 | 1.630 | 0.144 | 0.145 | 0.143 | 2.25/2.30 | — | 2.25/2.30 | 18 | — | 18 | Group 20 | 50 | 100 |
| 2324-39 (Repr.) | 319-54 | 1.610 | 0.148 | 0.155 | 0.144 | 2.250/2.275 | — | 2.275 | 16 | — | 18 | Group 21 | 70 (ent.)<br>50 (exit) | 210 |
| 2324-40 | 002-2 | 1.796 | 0.100 | 0.111 | 0.107 | 2.275/2.300 | — | 2.300 | 19 | — | 21 | Group 21 | 50 (ent.)<br>35 (exit) | 100 |
| 2334-41 | 002-4 | 1.711 | 0.140 | 0.154 | 0.142 | 2.275/2.300 | — | 2.300/2.325 | 19 | — | 23 | Group 21 | 55 (ent.)<br>45 (exit) | 100 |
| 2334-42 (0007) | 002-7 | 1.724 | 0.129 | 0.148 | 0.144 | 2.275/2.300 | — | 2.300/2.325 | 19 | — | 23 | Group 11 | 55 (ent.)<br>35 (exit) | 100 |
| 2322-43 | 002-10 | 1.788 | 0.092 | 0.116 | 0.114 | 2.35/2.40 | — | 2.35/2.40 | 30 | — | 30 | Group 20 - Ent.<br>Group 10 - Exit | 80 (ent.)<br>20 (exit) | 000 |
| 2324-44 (0008) | 002-12 | 1.738 | 0.135 | 0.141 | 0.136 | 2.275/2.300 | — | 2.275/2.300 | 19 | — | 19 | Group 11 - Ent.<br>Group 10 - Exit | 30 (ent.)<br>15 (exit) | 000 |
| 2424-1 | 319-47 | 1.730 | 0.080 | 0.140 | 0.138 | 2.25/2.30 | 2.30 | 2.25/2.30 | 18 | 21 | 18 | Group 20 | 50 (ent.)<br>40 (exit) | 210 |
| 2424-1 | 319-47 | 1.730 | 0.080 | 0.140 | 0.138 | 2.25/2.30 | 2.30 | 2.25/2.30 | 18 | 21 | 18 | Group 20 | 50 (ent.)<br>40 (exit) | 210 |
| 2422-2 (Repr.) | 319-49 | 1.810 | 0.078 | 0.105 | 0.079 | 2.25 | — | 2.25 | 15 | — | 15 | Group 21 | 65 (ent.)<br>50 (exit) | 210 |
| 2424-3 (Repr.) | 319-51 | 1.755 | 0.117 | 0.132 | 0.115 | 2.25/2.30 | — | 2.25/2.30 | 18 | — | 18 | Group 21 - Ent.<br>Group 20 - Exit | 90 (ent.)<br>50 (exit) | 200 |

## Table XV. 1.72-inch HIPPO Throat Insert Data Summary (fired nozzles only).

| Ident. No. | Deposition Run No. | Total Gas Flow Rate (SCFH) | Volume Percent $CH_4$ | Volume Percent MTS | Deposition Temperature (°F) | Deposition Rate (mils/hr) At Entrance | Deposition Rate (mils/hr) At Throat | Deposition Rate (mils/hr) At Exit | Micro Code | SiC Percentage At Entrance | SiC Percentage At Exit |
|---|---|---|---|---|---|---|---|---|---|---|---|
| 2322-9 0001 | 033-40 | 164.2 | 2.44 | 0.38 | 3200 | 20.4 | 22.7 | 21.1 | 21 | 18 | 24 |
| 2322-18 0002 | 033-65 | 164.6 | 2.43 | 0.34 | 3200 | 16.2 | 20.6 | 19.8 | 20 | 21 | 19 |
| 2332-23 0003 | 319-11 | 164.6 | 2.43 | 0.36 | 3200 | 16.2 | 21.1 | 19.2 | 30/20 | 21 | 23 |
| 2334-27 0004 | 319-20 | 244.7 | 1.63 | 0.29 | 3200 | 19.0 | 20.7 | 19.2 | 20 | 24 | 24 |
| 2324-35 0005 | 319-46 | 243.6 | 1.64 | 0.24 | 3200 | 17.7 | 19.9 | 18.6 | 20 | 18 | 18 |
| 2324-37 0006 | 319-50 | 243.6 | 1.64 | 0.24 | 3200 | 18.6 | 19.3 | 18.2 | 20 | 18 | 21 |
| 2334-42 0007 | 002-7 | 243.5 | 1.64 | 0.22 | 3200 | 15.3 | 17.5 | 17.0 | 11 | 19 | 23 |
| 2324-44 0008 | 002-12 | 243.6 | 1.64 | 0.23 | 3200 | 16.0 | 16.7 | 16.1 | 11/10 | 19 | 19 |
| | | | | | AVERAGES | 17.4 | 19.8 | 19.0 | | 20 | 21 |
| | | | | | STD. DEV. | | | | | 2 | 2 |

Substrate dimensions were recorded before and after coating to check for potential coating/substrate CTE mismatch. The data are shown in Table XVI. No significant change is apparent.

The first four 1.72-inch deposition runs were conducted in the 6-inch, induction heated, carbon tube furnace. Only one of these, 2322-2, yielded an acceptable coating. This part was sectioned axially to determine the SiC concentration in the throat area which was found to be acceptable at 18 percent.

After the fourth 1.72-inch fabrication run, the deposition process was shifted to the Pereny 4-inch carbon tube resistance furnace. At this time the deposition temperature was reduced to 3,200°F which was considered to be nominal in the 4-inch furnace. Also, the process nitrogen gas flow rate was changed to and fixed at 150 SCFH.

The first two 1.72-inch deposition runs, in the 4-inch furnace, were conducted using identical deposition parameters. The first of these, 2332-5, utilized Dow $CH_3SiCl_3$ as the SiC source and the second, 2322-6, Union Carbide $CH_3SiCl_3$. Both parts were sectioned axially and examined. The coating at the entrance end of 2332-5 was 6 percent higher in SiC concentration and less thick than 2322-6.

Indications were that Union Carbide $CH_3SiCl_3$ was equal to and possibly superior to the Dow material when depositing on this configuration. At this time Union Carbide $CH_3SiCl_3$ was selected as the SiC source material and was utilized for all subsequent deposition runs in the program.

The next four deposition runs, 2322-7 through 2322-10, were all conducted utilizing similar deposition conditions except that the injector to throat spacing was reduced to 5 inches. The entrance/throat coating thickness ratio as well as the entrance/exit%SiC ratio in these four runs was inconsistent. This thickness ratio inconsistency continued through to Run 2324-26, although the composition became consistent. At the same time, for some inexplicable reason, the SiC needle size rose to code 30. At Run 2324-26 the injector spacing was changed to 6 inches. After this the entrance/throat ratio never fell below 0.75/1 and averaged 0.90/1.

For deposition run, 2334-27, the process nitrogen flow rate was increased from 150 to 230 SCFH to improve the coating surface texture. This corresponded to a calculated linear flow rate of reactants, at the throat, of 20 and 29 feet per second, respectively. The coating surface texture was improved. However, it was difficult to maintain this rate using the existing flowmeter, and to alleviate this problem, the next six runs, 2324-29 through 2324-33, were conducted using 210 SCFH. Starting with Run 2324-34 a new flowmeter tube was installed and a process nitrogen rate of 230 SCFH was utilized for all subsequent 1.72-inch deposition runs.

The increase in process nitrogen rate did not immediately solve the surface roughness problem. Much of this turned out to be the result of poor technique in cleaning and assembling the deposition hardware. To assure the best possible coating texture requires that the following procedure must be followed scrupulously:

a. All mating diameters in the deposition chamber should be concentric within 0.010 inch.

b. All of the deposition fixturing should be wiped with a dry, lint-free cloth and blown with dry nitrogen before assembly.

c. After assembly the deposition fixturing should be blown out, again to remove dust generated during assembly.

Table XVI. 1.72-Inch HIPPO Throat Insert Substrate Dimensional Data.

| Ident. No. | Deposition Run No. | Substrate Material | Percent Change in Substrate Dimensions After Coating | | | |
|---|---|---|---|---|---|---|
| | | | Entrance OD | Mid OD | Exit OD | Length |
| 2322-8 | 033-39 | ATJ/A | 0 | 0 | 0 | 0 |
| 2322-9 0001 | 033-40 | ATJ | 0 | -0.10 | 0 | 0 |
| 2322-10 | 033-41 | ATJ | 0 | 0 | 0 | - |
| 2322-11 | 033-45 | ATJ | -0.03 | -0.03 | -0.03 | 0 |
| 2332-12 | 033-47 | ATJ | +0.03 | +0.03 | 0 | -0.06 |
| 2332-13 | 033-48 | ATJ | 0 | 0 | -0.03 | -0.06 |
| 2322-14 | 033-49 | ATJ | +0.03 | 0 | +0.03 | +0.06 |
| 2332-15 | 033-51 | ATJ | 0 | +0.03 | 0 | 0 |
| 2322-16 | 033-62 | ATJ | 0 | -0.03 | 0 | -0.06 |
| 2332-17 | 033-63 | ATJ | -0.03 | -0.03 | -0.03 | - |
| 2322-18 0002 | 033-65 | ATJ | 0 | 0 | +0.03 | 0 |
| 2322-19 | 319-1 | ATJ | 0 | -0.03 | +0.03 | 0 |
| 2322-20 | 319-8 | ATJ | -0.03 | 0 | 0 | 0 |
| 2322-21 | 319-9 | ATJ | -0.03 | 0 | 0 | -0.06 |
| 2322-22 | 319-10 | ATJ | 0 | 0 | -0.03 | +0.06 |

Table XVI. (continued).

| Ident. No. | Deposition Run No. | Substrate Material | Percent Change in Substrate Dimensions After Coating | | | |
|---|---|---|---|---|---|---|
| | | | Entrance OD | Mid OD | Exit OD | Length |
| 2332-23 0003 | 319-11 | ATJ | -0.03 | 0 | +0.03 | 0 |
| 2322-24 | 319-12 | ATJ | +0.03 | 0 | 0 | -0.06 |
| 2334-25 | 319-17 | ATJ | +0.03 | +0.03 | 0 | +0.06 |
| 2324-26 | 319-19 | ATJ | 0 | 0 | +0.06 | 0 |
| 2334-27 0004 | 319-20 | ATJ | +0.03 | 0 | +0.06 | - |
| 2334-28 | 319-24 | ATJ | -0.03 | 0 | 0 | 0 |
| 2324-29 | 319-33 | ATJ | -0.03 | -0.03 | +0.03 | 0 |
| 2324-31 | 319-42 | ATJ | -0.03 | -0.03 | -0.03 | -0.06 |
| 2324-32 | 319-43 | ATJ | -0.03 | -0.03 | -0.03 | -0.06 |
| 2324-33 | 319-44 | ATJ | -0.03 | -0.03 | -0.03 | 0 |
| 2324-34 | 319-45 | HLM | -0.07 | -0.03 | -0.03 | -0.06 |
| 2324-35 0005 | 319-46 | ATJ | +0.03 | 0 | +0.07 | -0.06 |
| 2324-37 0006 | 319-50 | ATJ | -0.03 | -0.06 | -0.03 | -0.06 |
| 2324-40 | 002-2 | ATJ | -0.03 | +0.06 | +0.03 | +0.06 |

Table XVI. (continued).

| Ident. No. | Deposition Run No. | Substrate Material | Percent Change in Substrate Dimensions After Coating | | | |
|---|---|---|---|---|---|---|
| | | | Entrance OD | Mid OD | Exit OD | Length |
| 2334-41 | 002-4 | ATJ | -0.03 | 0 | -0.03 | -0.06 |
| 2334-42 0007 | 002-7 | ATJ | -0.03 | +0.10 | +0.10 | +0.06 |
| 2322-43 | 002-10 | ATJ | +0.13 | -0.03 | 0 | +0.06 |
| 2324-44[a] 0008 | 002-12 | ATJ | -0.42 | -0.39 | -0.29 | 0 |
| Average ATJ Dimensional Change | | | -0.01 | -0.01 | +0.01 | -0.01 |

[a] Measurements appear to be in error. Data not used.

d. Thoroughly clean and blow off injector before insertion into deposition assembly.

e. After deposition temperature is reached, blow out deposition chamber at maximum rate possible through injector.

f. Check exhaust tube periodically and clean if necessary.

Whenever this procedure was followed no serious problems were encountered with surface texture of the coating using the established deposition conditions.

No significant problems were encountered in the deposition process from Run 2324-35 to 2324-39. During this time inserts for Firings 0005 and 0006 and one reproducibility specimen, 2324-39, were fabricated.

During Run 2324-40, the throat deposition rate decreased from 19.5 mils/hr in previous runs to 17.8 mils/hr. This was apparently caused by a malfunction in the furnace which changed the temperature profile in the deposition chamber. This reduction in the deposition rate required an increase in the deposition time. Because of the increased deposition time more $CH_3SiCl_3$ had to be added to the bubbler. This filled the bubbler to capacity which caused droplets of $CH_3SiCl_3$ to become entrained in the gas stream during the initial stage of deposition. This caused large momentary increases in the $CH_3SiCl_3$ rate which formed thin bands of increased SiC concentration in the coating. These bands were so prominent in the coatings on 2324-40 and 2334-41 that they were considered to be unsuitable for firing. Run 2334-42 was acceptable for Firing 0007 and was the last 1.72-inch insert coated in the 4-inch Pereny furnace.

The last two standard 1.72-inch inserts were fabricated in the 6-inch Pereny furnace because the 4-inch furnace was inoperable. A definite change in the coating characteristics took place whenever furnaces were changed. The SiC phase was much finer and the surface condition was considerably less rough. This is probably related to differences in the longitudinal temperature profile of the deposition chambers.

The first run, 2322-43, conducted in the 6-inch furnace utilized exactly the same deposition parameters as the previous run which was conducted in the 4-inch furnace. The throat coating deposition rate dropped from 17.5 to 13.7 mils/hr and the SiC concentration increased from an average of 21 percent to an average of 30 percent. The entrance/throat coating thickness ratio fell from 0.87/1 to 0.79/1.

The next deposition run, 2324-44, was conducted with the injector spacing changed from 6 to 6-1/2 inches. All other parameters were the same as those of the previous run. Post deposition examination showed the throat coating deposition rate to have increased to 16.7 mils/hr and the SiC concentration to have fallen to 19 percent. The entrance/throat coating thickness ratio was 0.96/1. This part was accepted for the last HIPPO motor firing of the program.

Three 1.72-inch wraparound nozzle throat inserts were fabricated to determine the uniformity of the coating using this configuration. All three coatings were applied to PG precoated HLM substrates to facilitate removal of the coating from the substrate. These runs were conducted between standard 1.72-inch deposition Runs 2324-35 and 2324-38.

The first wraparound 1.72-inch insert, 2424-1, was fabricated using the same deposition conditions as the previous standard 1.72-inch insert deposition run. Post deposition examination showed the entrance/throat coating thickness ratio to be 0.57/1. This was somewhat less than desired.

The next two wraparound nozzle insert fabrication runs, 2442-2 and 2424-3, were conducted in the 6-inch Pereny furnace with the injector spacing increased to 6-3/4 inches. This increased the entrance/throat coating thickness ratio to 0.74/1 and 0.89/1, respectively. These two coatings were used in the coating reproducibility studies.

## 4. 1.72-INCH HIPPO ENTRANCE APPROACH SECTIONS

Twenty-seven 1.72-inch entrance (see Figure 15) approach section fabrication runs were conducted. The deposition assembly configuration is shown in Figure 16. The first 16 of these utilized the 6-inch induction heated furnace as the heat source. The remainder were conducted in the Pereny 6-inch resistance furnace.

Coating process conditions are shown in Table XVII and coating characterization data are shown in Table XVIII. Additional process and coating data, for the eight fired parts, are summarized in Table XIX.

A higher $CH_3SiCl_3$ flow rate was required to fabricate the 1.72-inch entrance approach sections than was required for the smaller parts fabricated. This necessitated the use of a much larger bubbler. The basic design of this bubbler was similar to that of the one used for smaller parts. Maximum capacity of this bubbler was five gallons. A broad range of $CH_3SiCl_3$ rates were used during this series of runs. A graph showing the $CH_3SiCl_3$ rate versus bubbler nitrogen flow rate is shown in Figure 17. This graph was used to determine the $CH_3SiCl_3$ rate during the first twelve runs when bubbler weights had not been recorded.

Three distinct microstructural variations were encountered during the fabrication of these parts.

The first microstructure group, Group A, was produced during the first sixteen runs when depositions were being conducted in the 6-inch induction furnace. Included in this first group are the entrance approach sections fired in HIPPO's 0001, 0002, 0003 and 0004. The coatings fabricated during this phase, contained moderate to large acicular SiC and were generally classified as groups 20, 21, 30 and occasionally as groups 60 and 70.

When the deposition proccess was shifted to the 6-inch resistance furnace the coating microstructure changed drastically. This resulted in the formation of Group B which are Runs 17 to 25 inclusive. The SiC phase was extremely fine and although frequenly classified in groups 10 and 11 it was much finer than that which is normally associated with these groups. On two occasions the coatings fell into the group 40 class. The deposition rate also showed a marked decline. Attempts were made to coarsen the SiC needles throughout the Group B series. However, this goal was never achieved until the temperature was raised in Group C. Two of the entrance approach sections fabricated during the Group B phase were utilized for HIPPO firings 0005 and 0006.

The third microstructure variation Group C, occurred during the last two runs, when the deposition temperature was increased to 3,500°F. The two parts were utilized in HIPPO firings 0007 and 0008. The microstructure of these parts was definitely in the group 11 to group 21 range; i.e., they contained coarser SiC needles than previously encountered with the 6-inch Pereny. The deposition rate increased markedly over that of the previous series and was even slightly above that of the first series of runs conducted in the 6-inch induction furnace.

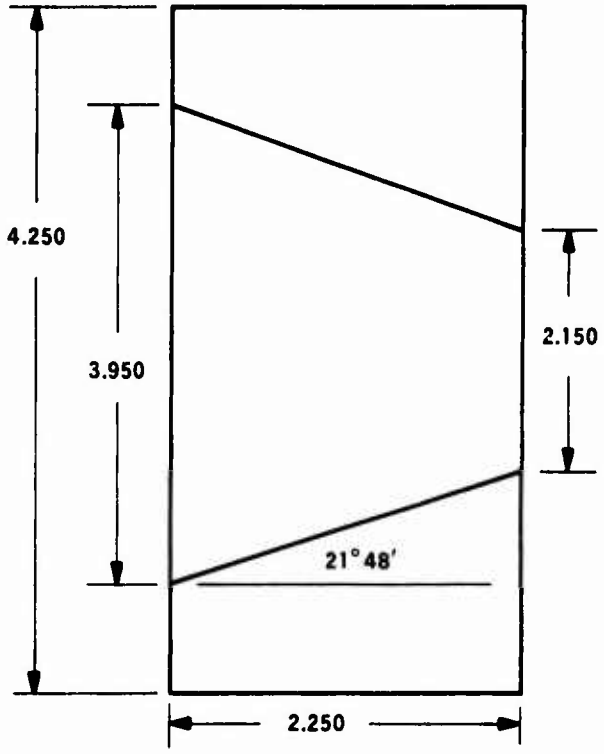



Figure 15. 1.72-Inch HIPPO Entrance Approach Section Substrate.

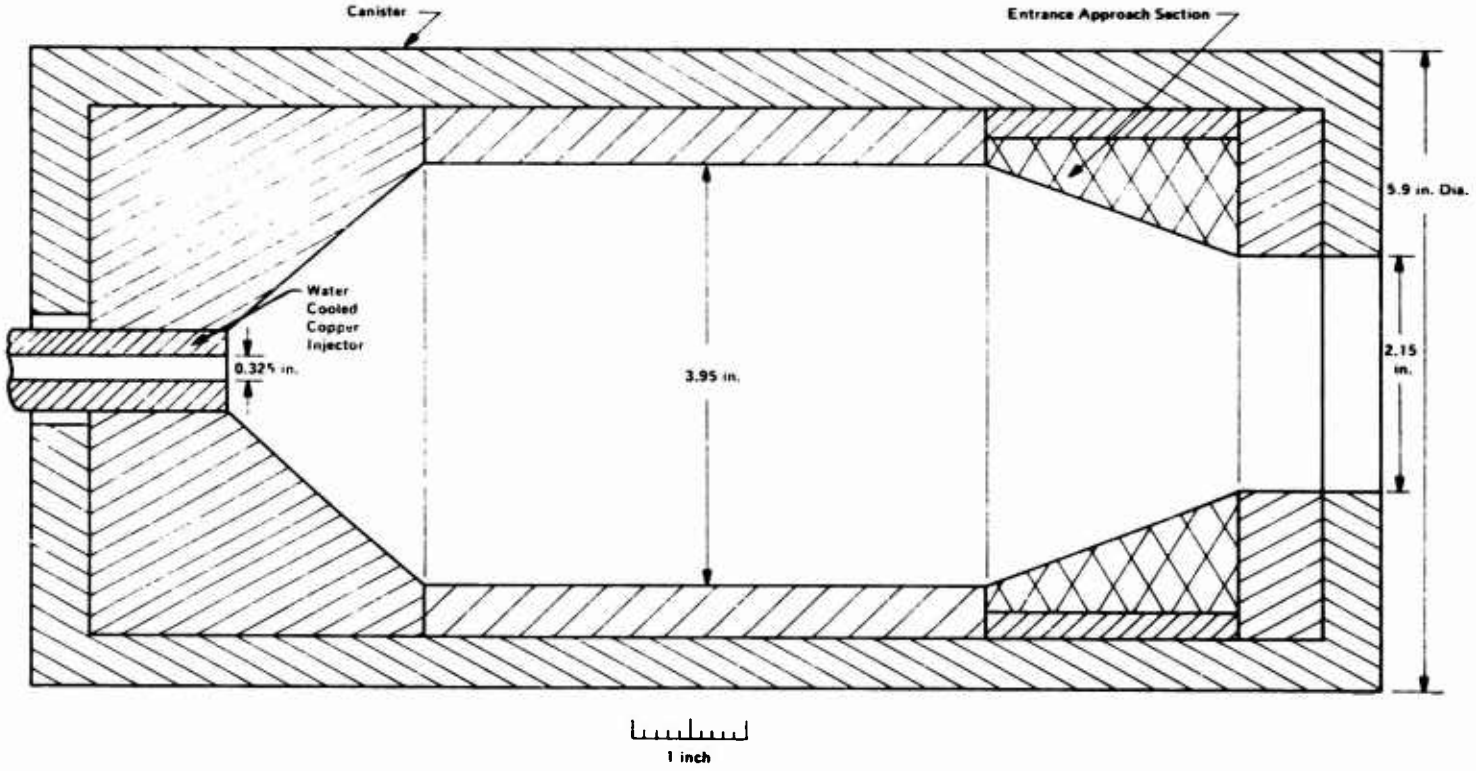



Figure 16. 1.72-inch HIPPO Entrance Approach Section Deposition Assembly.

## Table XVII. 1.72-inch HIPPO Entrance Approach Section Coating Process Conditions.

| Ident. No. | Deposition Run No. | Annulus $N_2$ (SCFH) | Process $N_2$ (SCFH) | Process $CH_4$ (SCFH) | Process $N_2$ to Bubbler (SCFH) | $CH_3SiCl_3$ Rate (g/hr) | Deposition Temperature (°F) | Deposition Time (hr) | Injector Spacing (in.) | Remarks |
|---|---|---|---|---|---|---|---|---|---|---|
| | | | | | | GROUP A | | | | |
| 2700-1 | 309-1 | 11.0 | 200 | 4.0 | 6 | 90[a] | 3,300 | 4.58 | 7 1/2 | Run conducted in 6-inch induction furnace. |
| 2700-2 | 309-2 | 11.0 | 400 | 8.0 | 6 | 90[a] | 3,300 | 6.00 | 10 | Process $N_2$ rate doubled to reduce dwell time of gas in chamber and therefore improve coating quality. |
| 2712-3 | 309-3 | 11.0 | 400 | 8.0 | 7 | 100[a] | 3,200 | 6.00 | 10 | Bubbler $N_2$ rate increased to increase SiC concentration. Deposition temperature reduced for same reason. |
| 2712-4 | 309-5 | 11.0 | 470 | 8.0 | 65 | 500[a] | 3,200 | 4.50 | 10 | Bubbler $N_2$ rate increased to increase SiC concentration. Process $N_2$ rate increased to improve coating microstructure. |
| 2730-5 | 309-6 | 11.0 | 350 | 8.0 | 65 | 500[a] | 3,200 | 4.50 | 10 | Process $N_2$ reduced to increase SiC crystal size. |
| 2732-6 (Repr) | 309-7 | 11.0 | 470 | 8.0 | 65 | 500[a] | 3,200 | 4.50 | 8 1/2 | 3-inch long tapered section added to entrance end of deposition chamber to reduce upstream turbulence. |
| 2732-7 | 309-8 | 11.0 | 470 | 8.0 | 60 | 480[a] | 3,200 | 4.50 | 8 1/2 | MTS flow started and stabilized before introduction into furnace. MTS flow introduced for 1 to 2 minutes prior to introducing $CH_4$. |
| 2732-8 (0001) | 309-9 | 11.0 | 470 | 8.0 | 54 | 450[a] | 3,200 | 4.50 | 8 1/2 | Same procedure as 309-8. $N_2$ to bubbler reduced to reduce SiC concentration. |
| 2741-9 | 309-12 | 11.0 | 705 | 8.0 | 60 | 480[a] | 3,000 | 6.00 | 8 1/2 | Process $N_2$ rate increased and deposition temperature reduced to determine their effect on the coating microstructure. |
| 2732-10 (0002) | 309-13 | 11.0 | 470 | 8.0 | 45 | 390[a] | 3,200 | 4.50 | 8 1/2 | Returned to parameters of 309-9 with exception of MTS rate which was reduced to reduce SiC concentration. |
| 2732-11 (Repr) | 309-16 | 11.0 | 470 | 8.0 | 54 | 450[a] | 3,200 | 4.50 | 8 1/2 | MTS rate increased to increase SiC concentration which was believed to be too low because of erroneous density fluids. |
| 2742-12 (0003) | 309-20 | 11.0 | 470 | 8.0 | 54 | 450[a] | 3,200 | 4.50 | 8 1/2 | None |
| 2743-13 | 309-22 | 11.0 | 470 | 8.0 | 54 | 428 | 3,200 | 6.00 | 8 1/2 | Deposition time increased to increase coating thickness to 150 mils. |
| 2734-14 | 309-24 | 11.0 | 470 | 8.0 | 54 | 454 | 3,200 | 7.00 | 8 1/2 | Same as above. |
| 2734-15 (0004) | 309-27 | 11.0 | 470 | 8.0 | 41 | 370[a] | 3,200 | 7.00 | 8 1/2 | MTS rate reduced to reduce SiC concentration. Deposition time not reduced because reduced MTS rate should reduce thickness. |
| 2734-16 | 309-30 | 11.0 | 470 | 8.0 | 27 | 272 | 3,200 | 7.0 | 8 1/2 | Same as above. |

[a]Derived from Figure

## Table XVII. (continued)

| Ident. No. | Deposition Run No. | Annulus $N_2$ (SCFH) | Process $N_2$ (SCFH) | Process $CH_4$ (SCFH) | Process $N_2$ to Bubbler (SCFH) | $CH_3SiCl_3$ Rate (g/hr) | Deposition Temperature (°F) | Deposition Time (hr) | Injector Spacing (in.) | Remarks |
|---|---|---|---|---|---|---|---|---|---|---|
| | | | | | | GROUP B | | | | |
| 2732-17 | 309-34 | 11.0 | 500 | 8.0 | 15 | 193 | 3,200 | 8.0 | 8 1/2 | New 6-inch Pereny resistance furnace used for first time. MTS rate reduced to lower SiC concentration. Injector orifice increased from .200 to .325. |
| 2734-18 (0006) | 309-43 | 15.0 | [illegible] | 10.0 | 15 | 153 | 3,240 | 10.0 | 8 1/2 | Increased $CH_4$ rate to increase PG content. Increased deposition time to increase coating thickness. |
| 2734-19 (0005) | 309-[illegible] | 15.0 | [illegible] | [illegible] | [illegible] | 198 | 4,220 | [illegible] | 9 | Injector spacing increased to increase SiC crystal size. MTS rate reduced to lower SiC concentration. |
| 2734-20 | 309-[illegible] | 15.0 | 500 | 10.0 | [illegible] | [illegible] | 3,230 | 10.[illegible] | 9 | Suspect MTS rate may be in error. |
| 2734-21 | 309-[illegible] | 15.0 | [illegible] | 10.0 | [illegible] | 170 | 3,20[illegible] | 10.[illegible] | 9 | Reduced process $N_2$ rate to that of earlier runs in attempt to increase SiC crystal size. |
| 2732-22 | 309-52 | 15.0 | [illegible] | 10.0 | [illegible] | 144 | [illegible]3[illegible]0 | 10.[illegible] | 9 | Temperature increased to increase SiC crystal size. |
| 2734-23 | 309-54 | 15.0 | [illegible] | 10.0 | [illegible] | 142 | [illegible]2[illegible]0 | 10.[illegible] | 9 | Process $N_2$ rate decreased to increase gas temperature before it reaches coating surface, in attempt to increase SiC crystal size. |
| 2732-24 | 309-56 | 15.0 | 470 | 10.0 | [illegible] | [illegible] | 3,290 | 10.0 | 9 | Injector orifice diameter reduced from 0.325 to 0.200 to increase turbulence and, hopefully, increase SiC crystal size. |
| 2724-25 | 002-1 | 15.0 | [illegible] | 15.0 | 11 | 14[illegible] | 3,300 | 8.0 | 9 | $CH_4$ rate and MTS rate both increased to increase deposition rate and increase SiC crystal size. Injector orifice 0.325 inch. |
| | | | | | | GROUP C | | | | |
| 2726-26 (0008) | 002-3 | 15.0 | 470 | 15.0 | 15 | 2[illegible]4 | 3,550 | 9.0 | 9 | Temperature increased drastically to increase SiC crystal size. MTS rate increased to compensate for expected SiC concentration loss. |
| 2724-27 (0007) | 002-6 | 15.0 | 470 | 15.0 | 22 | 236 | 3,500 | 4.8 | 9 | Deposition time reduced to reduce coating thickness. MTS rate increased to increase SiC concentration. |

[a] Derived from Figure 17.

## Table XVIII. 1.72-inch HIPPO Entrance Approach Section Coating Evaluation Data.

| Ident. No. | Deposition Run No. | Coating Thickness (in.) | | Density (gm/cc) | | SiC[a] Concentration (%) | | Appearance | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | | | | | | | | Entrance | | | Exit | | |
| | | Entrance | Exit | Entrance | Exit | Entrance | Exit | Micro-Structure | Cone Angle (degree) | Surface Condition | Micro-Structure | Cone Angle (degree) | Surface Condition |
| GROUP A | | | | | | | | | | | | | |
| 2700-1 | 309-1 | 0.021 | 0.013 | 2.15 | 2.15 | 0 | 0 | Group 70 | — | Covered with soft growths | Group 70 | — | — |
| 2700-2 | 309-2 | 0.018 | 0.028 | 2.00 | 2.15/2.20 | 0 | 3 | Group 70 | — | 100 | Group 70 | — | 100 |
| 2712-3 | 309-3 | 0.074 | 0.092 | 2.20 | 2.20 | 6 | 6 | Not visible at 600X | — | 000 | Not visible at 600X | — | 000 |
| 2712-4 | 309-5 | 0.074 | 0.101 | 2.15/2.20 | 2.15/2.20 | 1 | 1 | SiC phase not visible at 1000X | 40 - 55 | 231 | SiC phase not visible at 1000X | 40 - 55 | 231 |
| 2730-5 | 309-6 | — | 0.020/0.050 | — | 2.35/2.40 | — | 30 | No coating present. Rough growths present around perimeter. Not micropolished at this end. | | | Coating rough and scalloped. Not micropolished. | | |
| 2732-6 (Repr) | 309-7 | 0.046 | 0.096 | 2.275/2.300 | 2.325 | 19 | 24 | Thin unalloyed PG band visible at interface at both ends. | | | | | |
| | | | | | | | | Group 30 | 55 | 200 | Group 21 | 35 | 200 |
| 2732-7 | 309-8 | 0.069 | 0.109 | — | 2.35/2.40 | — | 30 | Coating covered with nodules resulting from melting of the exit cleanout tube onto the substrate. Coating ends not micropolished. | | | | | |
| 2732-8 (0001) | 309-9 | 0.056 | 0.098 | 2.300/2.325 | 2.35/2.40 | 22 | 30 | Group 30 | 55 | 210 | Group 21 | 30 | 210 |
| 2741-9 | 309-12 | 0.055 | 0.062 | 2.45/2.50 | 2.60 | 40 | 52 | Group 21 | 25 - 55 | 000 | Group 20 | 25 - 35 | 000 |
| 2732-10 (0002) | 309-13 | 0.065 | 0.096 | 2.300/2.325 | 2.40 | 22 | 32 | Group 30 | 35 - 55 | 210 | Group 21 | 25 - 45 | 210 |
| 2732-11 (Repr) | 309-16 | 0.093 | 0.112 | 2.300 | 2.325 | 21 | 24 | Group 60 | 25 - 75 | 210 | Group 30 | 25 - 45 | 210 |
| 2742-12 (0003) | 309-20 | 0.074 | 0.101 | 2.30 | 2.55 | 21 | 47 | Group 60 | 45 - 65 (50 avg.) | 210 | Group 30 | 45 - 65 (50 avg.) | 010 |
| 2741-13 | 309-22 | 0.109 | 0.113 | 2.35/2.40 | 2.45/2.50 | 30 | 40 | Group 30 | 40 - 50 | 211 | Group 20 | 40 - 50 | 010 |
| 2734-14 | 309-24 | 0.134 | 0.161 | 2.35 | 2.45 | 27 | 37 | Group 60 | 50 - 70 | 011 | Group 60 | 50 - 70 | 011 |
| 2734-15 (0004) | 309-27 | 0.111 | 0.166 | 2.325/2.350 | 2.40/2.45 | 25 | 34 | Group 30 | — | 110 | Group 30 | — | 000 |
| 2734-16 | 309-30 | 0.111 | 0.138 | 2.30/2.35 | 2.40 | 24 | 32 | Group 20 | 40 - 60 (50 avg.) | 121 | Group 20 | 40 - 60 (50 avg.) | 121 |
| GROUP B | | | | | | | | | | | | | |
| 2732-17 | 309-34 | 0.080 | 0.092 | 2.30/2.35 | 2.40/2.45 | 24 | 34 | Group 11 | 60 | 100 | Group 10 Coating shows banding | 60 | 100 |
| 2734-18 (0006) | 309-40 | 0.108 | 0.152 | 2.25 | 2.35/2.40 | 15 | 30 | Group 20 | 75 | 100 | Group 10 | 55 | 000 |
| 2734-19 (0005) | 309-42 | 0.120 | 0.140 | 2.25 | 2.30/2.35 | 15 | 24 | Group 10 | 50 | 000 | Group 10 | 50 | 000 |
| 2724-20 | 309-46 | 0.120 | 0.148 | 2.25 | 2.275/2.300 | 15 | 19 | Group 40 | 55 | 100 | Group 40 | 55 | 000 |
| 2734-21 | 309-49 | 0.107 | 0.142 | 2.225/2.250 | 2.325/2.350 | 13 | 25 | Group 10 | 60 | 010 | Group 10 | 60 | 010 |
| 2732-22 | 309-52 | 0.100 | 0.118 | 2.250/2.275 | 2.300/2.325 | 16 | 22 | Group 40 | 50 | 010 | Group 40 | 50 | 010 |
| 2734-23 | 309-54 | 0.120 | 0.140 | 2.200/2.250 | 2.325/2.350 | 12 | 25 | Group 11 Coating contains 10, end to end, axial cracks. | 85 | 100 | Group 11 | 85 | 000 |
| 2732-24 | 309-56 | 0.072 | 0.076 | 2.350 2.400 | 2.350 2.400 | 30 | 30 | Group 11 | 65 | 000 | Group 11 | 65 | 000 |
| 2724-25 | 002-1 | 0.137 | 0.137 | 2.20 | 2.275 | 7 | 18 | Group 10 Coating contains 1, end to end, axial crack. | 45 | 000 | Group 10 | 20 | 000 |
| GROUP C | | | | | | | | | | | | | |
| 2726-26 (0008) | 002-3 | 0.206 | 0.241 | 2.225 | 2.250/2.275 | 12 | 16 | Group 11 | 40 | 200 | Group 11 | 45 | 000 |
| 2724-27 (0007) | 002-4 | 0.116 | 0.152 | 2.225 | 2.250 | 12 | 15 | Group 21 | 60 | 100 | Group 11 | 50 | 000 |

[a]From density weight percent SiC relationship.

Table XIX. 1.72-inch HIPPO Entrance Approach Section Data Summary (fired nozzles only).

| Ident. No. | Deposition Run No. | Total Gas Flow Rate (SCFH) | Volume Percent $CH_4$ | Volume Percent MTS | Deposition Temperature (°F) | Deposition Rate (mils/hr) At Entrance | Deposition Rate (mils/hr) At Exit | Micro Code | SiC Percentage At Entrance | SiC Percentage At Exit |
|---|---|---|---|---|---|---|---|---|---|---|
| GROUP A | | | | | | | | | | |
| 2732-8 0001 | 309-9 | 545 | 1.47 | 0.46 | 3200 | 12.5 | 21.8 | 30/21 | 22 | 30 |
| 2732-10 0002 | 309-13 | 536 | 1.49 | 0.41 | 3200 | 14.4 | 21.3 | 30/21 | 22 | 32 |
| 2742-12 0003 | 309-20 | 545 | 1.47 | 0.46 | 3200 | 16.5 | 22.4 | 60/30 | 21 | 47 |
| 2734-15 0004 | 309-27 | 532 | 1.51 | 0.38 | 3200 | 15.9 | 23.7 | 30/30 | 25 | 34 |
| | | | | | GROUP A - AVERAGES | 15.1 | 22.3 | 30 | 23 | 35 |
| GROUP B | | | | | | | | | | |
| 2734-19 0005 | 309-42 | 537 | 1.86 | 0.12 | 3200 | 12.0 | 14.0 | 10/10 | 15 | 24 |
| 2734-18 0006 | 309-40 | 541 | 1.85 | 0.16 | 3200 | 10.8 | 15.2 | 20/10 | 15 | 30 |
| | | | | | GROUP B - AVERAGES | 11.4 | 14.6 | 10 | 15 | 27 |
| GROUP C | | | | | | | | | | |
| 2724-27 0007 | 002-6 | 523 | 2.87 | 0.26 | 3500 | 24.2 | 31.7 | 21/11 | 12 | 15 |
| 2726-26 0008 | 002-3 | 516 | 2.91 | 0.24 | 3500 | 22.9 | 26.8 | 11/11 | 12 | 16 |
| | | | | | GROUP C - AVERAGES | 23.6 | 29.2 | 11 | 12 | 16 |

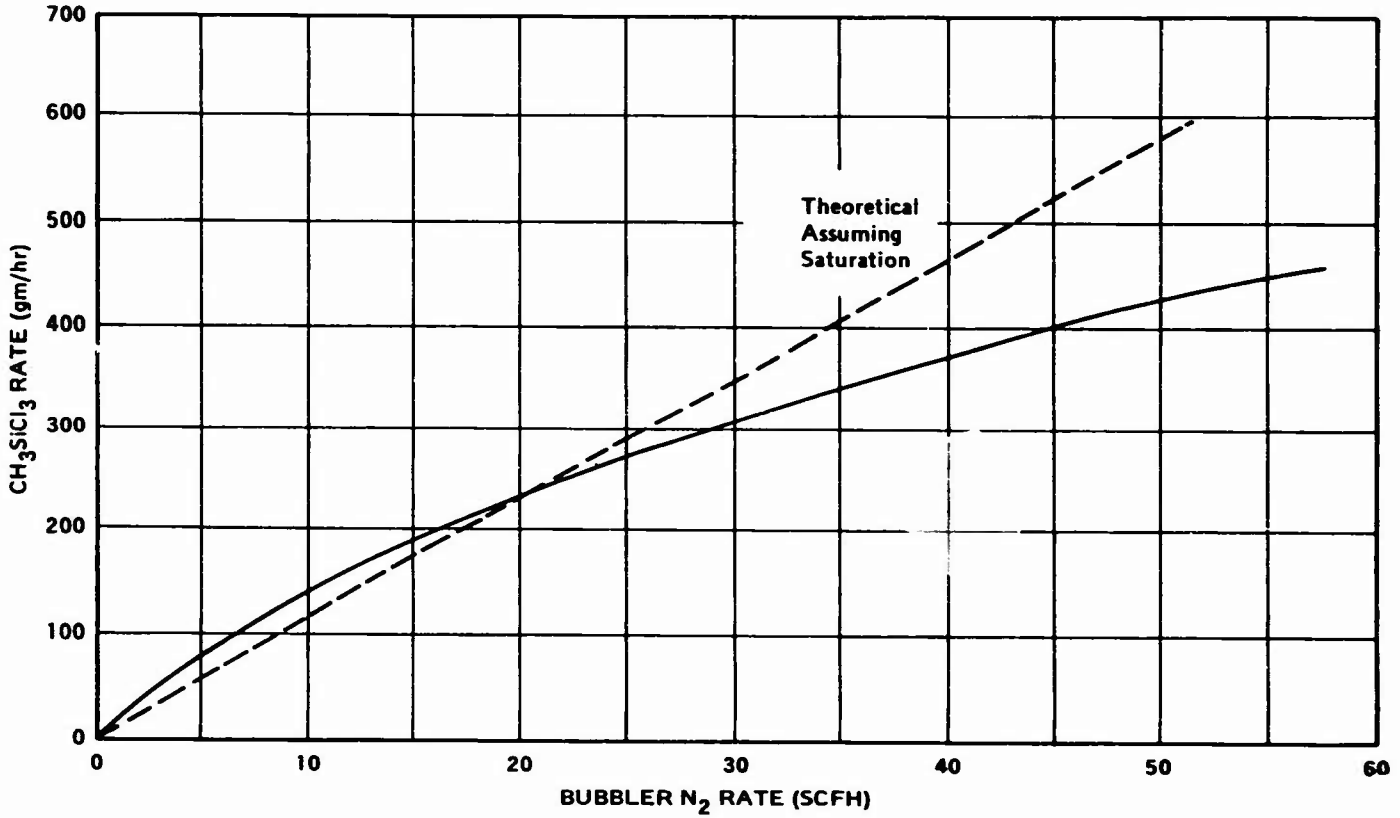



Figure 17. Bubbler $N_2$ Rate Versus $CH_3SiCl_3$ Rate (for 5-gallon bubbler operating at 50 psig and 70°F).

It appears that these variations in microstructure occur because of differences in the lateral thermal profile of the two deposition furnaces. It would at first appear that the deposition temperature of the surface being coated varies. However, since the temperature is read at the backside of the deposition canister and both canisters were of the same size, it is highly unlikely that variations in the order of 300°F could occur. Furthermore, periodic optical pyrometer calibrations revealed no large anomalies.

From the first deposition run, 2700-1, to the fourteenth, 2734-14, Dow $CH_3SiCl_3$ was used. All subsequent runs utilized Union Carbide $CH_3SiCl_3$. The $CH_3SiCl_3$ flow rate during the early runs was considerably higher than that which was later established as nominal. These high rates were though to be necessary to achieve a nominal 15 percent SiC containing coating. However, the density fluids, being used to determine the SiC concentration, were found to be in error and, as a result, the indicated SiC concentrations were too low. When this error was discovered the $CH_3SiCl_3$ rate was reduced to bring the SiC concentration nearer to the nominal 15 percent desired. It is interesting to note that coatings with SiC concentrations as high as 52 percent were fabricated without cooldown cracks. Flawed coatings were encountered, later in the program (Run 2734-23) at much lower concentrations, indicating that SiC concentration is not the only factor influencing the CTE of the coating.

Starting with Run 2734-15 Union Carbide $CH_3SiCl_3$ was utilized as the SiC source.

All of the substrates coated were fabricated from ATJ graphite with the axis normal to the grain direction.

Substrate dimensions were recorded before and after coating to check for potential coating/substrate CTE mismatch. The data are recorded in Table XX. A reduction in the substrate outside diameter occurred in nearly all of the parts coated. The data were also grouped according to SiC concentration to see if any difference in deformation occurs between high and low concentration of SiC. No significant difference was apparent.

The reduction in substrate diameter, which occurred, reinforces the characterization data which indicate that the CTE of the coating is higher than that of the ATJ substrate. Because of this mismatch cooldown stresses were, in some cases, high enough to cause coating failure as occurred in 2734-23 and 2724-25.

An average increase in the substrate length was indicated, as expected.

The summarized information on the eight coating runs that were used in the test firings is divided into the three groups, A, B and C in Table XIX. Again, there is no obvious reason why the products of Group A and Group B are so different except that a relationship may exist between the SiC crystal size and the MTS concentration in the reactant gas. It may also be caused by differing thermal gradients in different furnaces. It should be emphasized that the control of codeposit properties as a function of deposition parameters is still not well understood.

Table XX. 1.72-Inch HIPPO Entrance Approach Section Substrate Dimensional Data.

| Ident. No. | Deposition Run No. | Substrate Material | Percent Change in Substrate Dimensions After Coating: Entrance OD | Mid OD | Exit OD | Length | Comment |
|---|---|---|---|---|---|---|---|
| | | | | GROUP A | | | |
| 2700-2 | 309-2 | ATJ | +0.12 | +0.12 | +0.12 | -0.11 | <3 percent SiC concentration. Disregard. |
| 2712-3 | 309-3 | ATJ | -0.12 | -0.07 | -0.07 | - | 6 percent SiC concentration. Disregard. |
| 2712-4 | 309-5 | ATJ | +0.16 | +0.19 | +0.21 | - | 3 percent SiC concentration. Disregard. |
| 2732-6 | 309-7 | ATJ | -0.12 | -0.12 | -0.12 | 0 | |
| 2732-8 0001 | 309-9 | ATJ | -0.09 | -0.09 | -0.09 | +0.09 | |
| 2741-9 | 309-12 | ATJ | -0.05 | -0.05 | -0.05 | 0 | |
| 2732-10 0002 | 309-13 | ATJ | 0 | -0.05 | -0.02 | 0 | |
| 2732-11 | 309-16 | ATJ | +0.14 | +0.05 | +0.01 | 0 | Measurements appear to be in error. Disregard. |
| 2742-12 0003 | 309-20 | ATJ | -0.14 | -0.09 | -0.05 | +0.18 | |
| 2743-13 | 309-22 | ATJ | -0.05 | -0.02 | -0.02 | +0.13 | |
| 2734-14 | 309-24 | ATJ | -0.07 | -0.05 | -0.02 | +0.18 | |
| 2734-15 0004 | 309-27 | ATJ | -0.05 | -0.02 | -0.02 | 0 | |

Table XX. (continued).

| Ident. No. | Deposition Run No. | Substrate Material | Percent Change in Substrate Dimensions After Coating | | | | Comment |
|---|---|---|---|---|---|---|---|
| | | | Entrance OD | Mid OD | Exit OD | Length | |
| 2734-16 | 309-30 | ATJ | -0.05 | -0.05 | -0.02 | - | |
| | | | | GROUP B | | | |
| 2732-17 | 309-34 | ATJ | -0.07 | -0.02 | -0.02 | - | |
| 2734-18 0006 | 309-40 | ATJ | -0.07 | -0.05 | -0.05 | +0.13 | |
| 2734-19 0005 | 309-42 | ATJ | -0.14 | -0.05 | -0.05 | - | |
| 2724-20 | 309-46 | ATJ | -0.16 | -0.09 | -0.05 | +0.18 | |
| 2734-21 | 309-49 | ATJ | -0.09 | -0.07 | -0.05 | - | |
| 2732-22 | 309-52 | ATJ | -0.14 | -0.14 | -0.07 | +0.09 | |
| 2734-23 | 309-54 | ATJ | -0.05 | -0.02 | -0.02 | - | Coating cracked axially. |
| 2732-24 | 309-56 | ATJ | -0.12 | -0.02 | +0.05 | - | |
| 2724-25 | 002-1 | ATJ | -0.12 | -0.07 | +0.02 | - | Coating cracked axially. |
| | | | | GROUP C | | | |
| 2726-26 0008 | 002-3 | ATJ | -0.09 | -0.05 | 0 | +0.22 | |

Table XX. (continued).

| Ident. No. | Deposition Run No. | Substrate Material | Percent Change in Substrate Dimensions After Coating: Entrance OD | Mid OD | Exit OD | Length | Comment |
|---|---|---|---|---|---|---|---|
| 2724-27 0007 | 002-6 | ATJ | -0.02 | -0.05 | 0 | - | |
| Average Dimensional Change: | | | | | | | |
| | | Group A | -0.07 | -0.06 | -0.04 | +0.07 | |
| | | Group B | -0.11 | -0.06 | -0.03 | +0.13 | |
| | | Group C | -0.06 | -0.05 | 0 | +0.22 | |
| | | 12-25% SiC | -0.09 | - | -0.04 | - | |
| | | >25% SiC | -0.07 | - | -0.03 | - | |
| | | Overall Average[a] | -0.09 | -0.06 | -0.04 | +0.13 | |
| | | Standard Deviation[a] | 0.04 | 0.03 | 0.04 | 0.07 | |

[a]Omitting 309-2, 3, 4, and 11.

## SECTION VII

## REPRODUCIBILITY STUDY

A study was undertaken to determine whether the coating characteristics, density, percent SiC, microstructure, and thickness were reasonably consistent in both the axial and the circumferential directions along the inserts. Two standard HIPPO throat inserts, two wraparound design HIPPO throat inserts and two HIPPO entrance approach sections were each sectioned at four axial locations in planes perpendicular to the axis. The specific axial locations are shown in Figures 18, 19, and 20. The results of the study are contained in Tables XXI, XXII, and XXIII.

One of the standard throat inserts was examined at four equispaced, 90 degree, circumferential locations at each axial location, as shown in Table XXIV. No unexpected anomalies occurred, except: (1) some inconsistencies in density around the periphery (Table XXIV) and (2) some inconsistencies in the relationship between SiC content and density, particularly in the case of Run 2732-6 in Table XXIII.

The SiC contents were determined by LUVAC, Incorporated, by a wet chemical method. The weighed sample was pulverized in a mortar and pestle so as to completely pass through a 200 mesh screen. The powder was fused with $Na_2CO_3$ in a platinum crucible and leached with dilute HCl. Perchloric acid was added and the silicon was precipitated as hydrated silicic acid by evaporation of the volatile constituents from the liquor. The silicic acid was converted to $SiO_2$ by ignition with a Bunsen burner. This product was weighed. HF was then added and the material was again evaporated to drive off the silicon as $SiF_4$. The residue was then weighed. The difference between the two weighings gave the $SiO_2$ weight and allowed calculation of the SiC content in accordance with the formula:

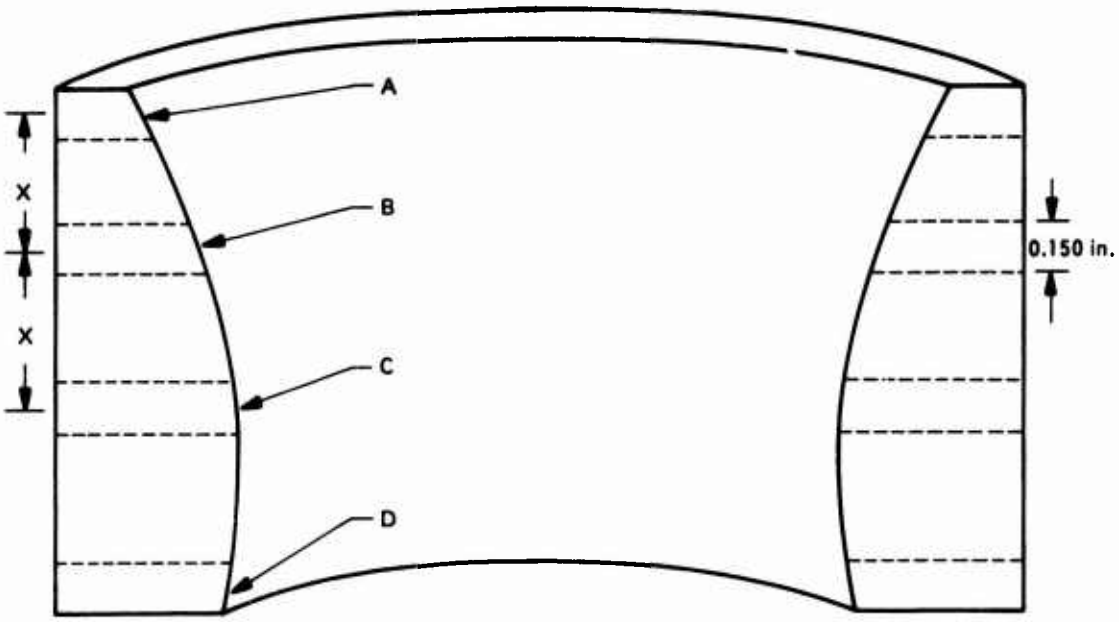
$$\% \text{ SiC} = \frac{\text{Weight of } SiO_2 \times 66.7}{\text{Weight of Sample}}$$

### 1. SiC CONTENT/DENSITY RELATIONSHIP

Using the SiC content data by wet chemistry from Luvac and the Atlantic Research density data for forty-one specimens from the reproducibility study containing between 15 and 25 percent SiC, the relationship between density and SiC content was established using a least squares equivalent technique and using the formula:

$$\frac{1}{d_{composite}} = \frac{\text{w/o SiC}}{321} + \frac{(100 - \text{w/o SiC})}{100\, d_G}$$

In this way, the density of the PG phase was determined to be 2.14 gm/cc, and the coefficient of variation was found to be 13 percent of the SiC content. The density of SiC was assumed to be 3.21 gm/cc. The standard deviation with respect to the PG phase density was found to be ±0.02 percent. The new plot of percent SiC versus density, based on the above formula is shown in Figure 21.




Enlarged to 2X Scale

Figure 18. HIPPO 1.72 Throat Insert Sectioning Plan.

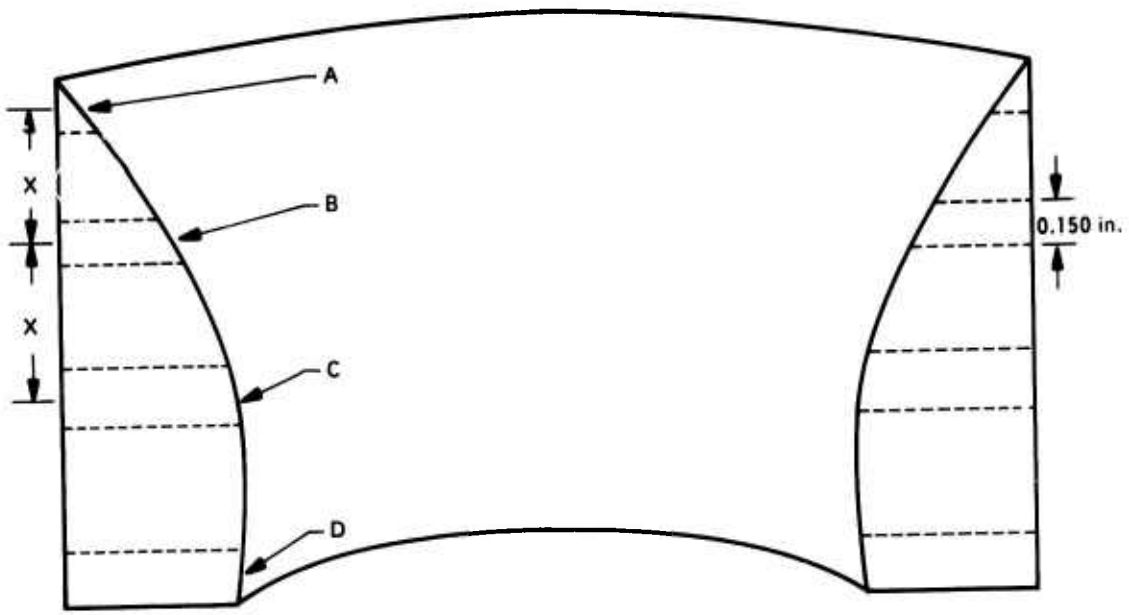



Figure 19. HIPPO 1.72 Wraparound Throat Insert Sectioning Plan.

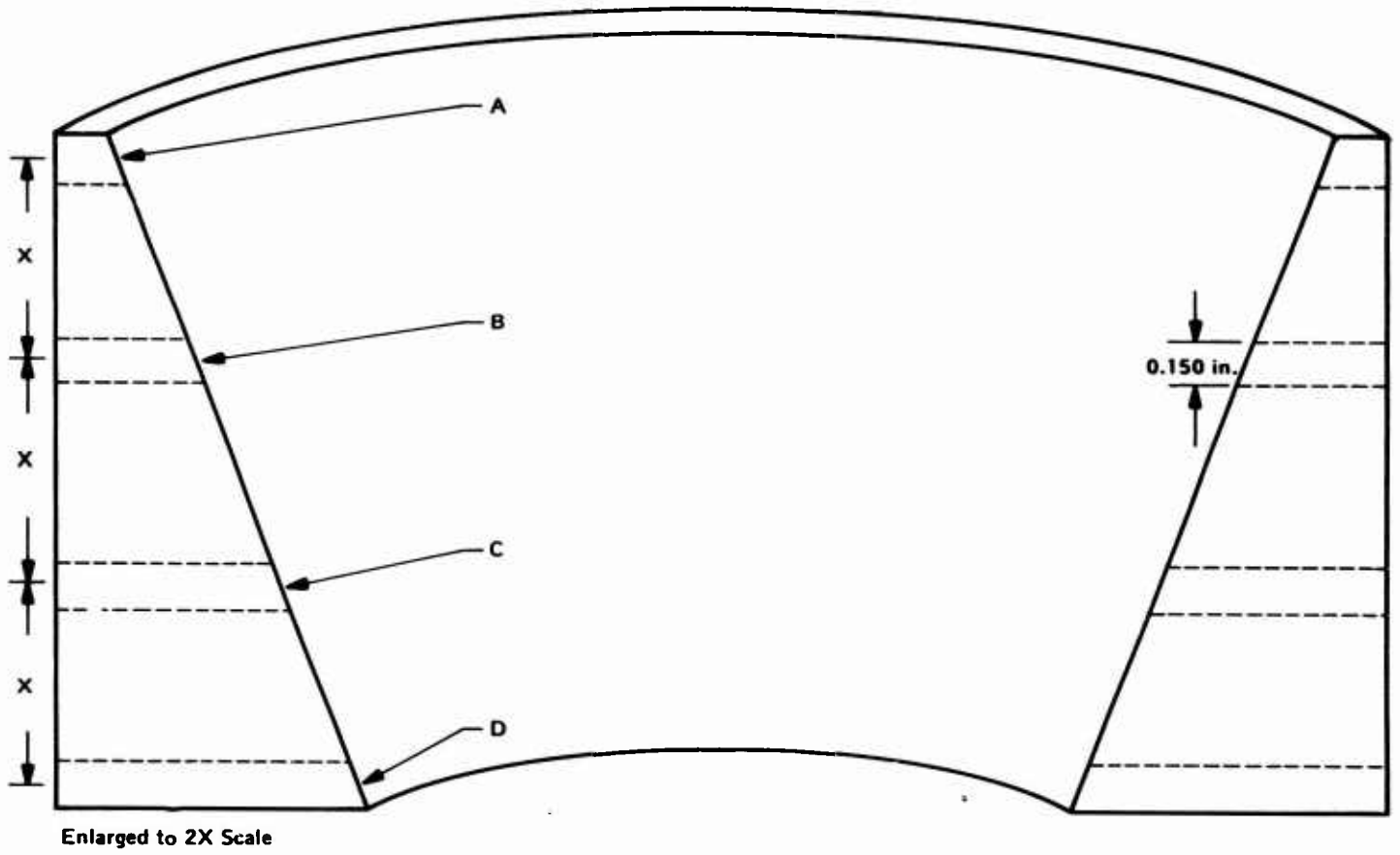



Figure 20. HIPPO 1.72 Entrance Approach Sectioning Plan.

Table XXI. HIPPO Throat Insert Reproducibility Part Shape 2324.

| | 2324-30 Run 319-41 | | | | 2324-39[a] Run 319-54 | | | |
|---|---|---|---|---|---|---|---|---|
| | Percent SiC | Density (g/cc) | Thickness (mils) | Micro Code | Percent SiC | Density (g/cc) | Thickness (mils) | Micro Code |
| Entrance | 21.17 | 2.275 | 158 | 21 | 16.80 | 2.26 | 144 | 11 |
| Halfway Between Entrance and Throat | 19.17 | 2.29 | 170 | 21 | 18.51 | 2.27 | 158 | 11 |
| Throat | 18.49 | 2.29 | 170 | 21 | 18.60 | 2.28 | 158 | 11 |
| Exit | 17.15 | 2.275 | 151 | 20 | 16.95 | 2.27 | 144 | 11 |

[a] Averages of 4 readings per Table XXIV

Table XXII. HIPPO Throat Insert Reproducibiiity Part Shape 2424 (wraparound).

| | 2422-2 Run 319-49 | | | | 2424-3 Run 319-51 | | | |
|---|---|---|---|---|---|---|---|---|
| | Percent SiC | Density (g/cc) | Thickness (mils) | Micro Code | Percent SiC | Density (g/cc) | Thickness (mils) | Micro Code |
| Entrance | 15.40 | 2.25 | 71 | 21 | 19.23 | 2.29 | 101 | 21 |
| Halfway Between Entrance and Throat | 17.62 | 2.26 | 102 | 21 | 22.04 | 2.31 | 147 | 21 |
| Throat | 18.13 | 2.26 | 90 | 21 | 15.93 | 2.31 | 136 | 21 |
| Exit | 16.79 | 2.26 | 79 | 21 | 20.90 | 2.30 | 118 | 21 |

Table XXIII. HIPPO Entrance Approach Section Reproducibility Part Shape 2722.

| | 2732-6 Run 309-7 | | | | 2732-11 Run 309-16 | | | |
|---|---|---|---|---|---|---|---|---|
| | Percent SiC | Density (g/cc) | Thickness (mils) | Micro Code | Percent SiC | Density (g/cc) | Thickness (mils) | Micro Code |
| Entrance | 18.65 | 2.25 | 59 | 70 | 18.60 | 2.31 | 98 | 70 |
| 1/3 from Entrance | 16.62 | 2.30 | 84 | 70 | 22.10 | 2.31 | 111 | 70 |
| 1/3 from Exit | 16.71 | 2.30 | 97 | 30 | 22.49 | 2.31 | 115 | 60 |
| Exit | 15.55 | 2.32 | 98 | 21 | 22.09 | 2.31 | 115 | 30 |

Table XXIV. HIPPO Throat Insert Reproducibility Part Shape 2324.

| | Circumferential Location | Percent SiC | Density (g/cc) | Thickness (mils) | Micro Code |
|---|---|---|---|---|---|
| Entrance | 0 | 17.17 | 2.26 | 144 | 11 |
| | 90 | 16.90 | 2.26 | 144 | 11 |
| | 180 | 17.20 | 2.26 | 144 | 11 |
| | 270 | 15.95 | 2.26 | 144 | 11 |
| AVERAGE | | 16.80 | 2.26 | 144 | 11 |
| Halfway Between Entrance and Throat | 0 | 18.95 | 2.275 | 158 | 11 |
| | 90 | 17.80 | 2.26 | 158 | 11 |
| | 180 | 18.32 | 2.275 | 158 | 11 |
| | 270 | 18.46 | 2.275 | 158 | 11 |
| AVERAGE | | 18.51 | 2.27 | 158 | 11 |
| Throat | 0 | 19.09 | 2.29 | 158 | 11 |
| | 90 | 18.17 | 2.26 | 158 | 11 |
| | 180 | 18.70 | 2.29 | 158 | 11 |
| | 270 | 18.46 | 2.29 | 158 | 11 |
| AVERAGE | | 18.60 | 2.28 | 158 | 11 |
| Exit | 0 | 18.63 | 2.26 | 144 | 11 |
| | 90 | 16.00 | 2.29 | 144 | 11 |
| | 180 | 17.93 | 2.26 | 144 | 11 |
| | 270 | 16.25 | 2.26 | 144 | 11 |
| AVERAGE | | 16.95 | 2.27 | 144 | 11 |

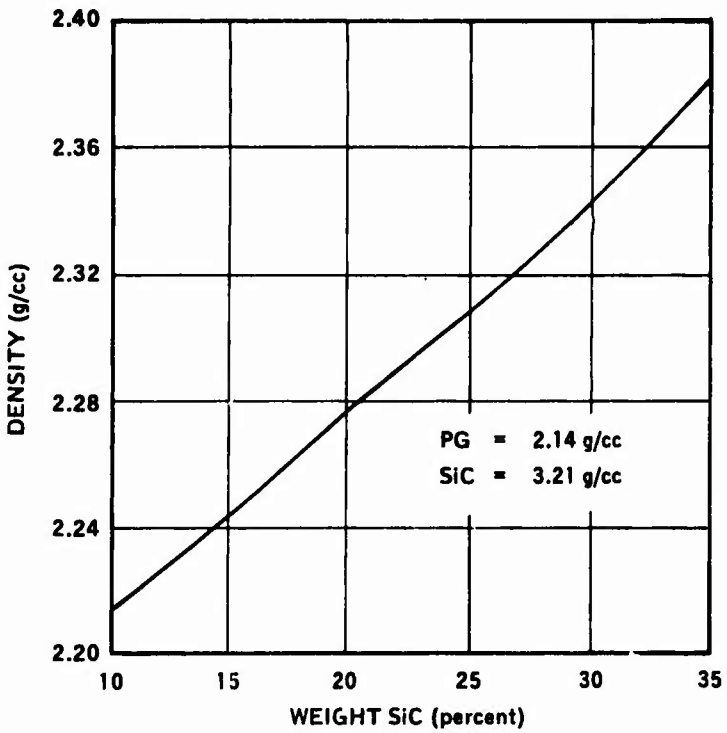



Figure 21. Relationship Between SiC Content and Density of PG/SiC Co-Deposit.

The raw data for the plot is contained in Section 8.0, Tables XXI through XXIII. The data are less reliable below 15 percent and above 25 percent. However, above 25 percent, the density relationship is considered to be more reliable than the ashing technique at Atlantic Research. The ashing technique tended to yield lower than true SiC contents, particularly at the higher SiC contents. This was undoubtedly caused by incomplete conversion of the SiC to $SiO_2$ which was probably due to inaccurate temperature control during ashing. This could probably be easily remedied. However, other proven techniques are now available. The density/%SiC relationship is recommended as adequate with an occasional spot check assay.

## SECTION VIII

## $SiCl_4$ AS SILICON SOURCE MATERIAL

A small effort was made to investigate the use of $SiCl_4$ as the silicon containing source material. It was felt that SiC could be deposited, from the $SiCl_4$, at higher temperatures than are attainable utilizing $CH_3SiCl_3$. The achievement of higher deposition temperatures would, hopefully, permit the formation of a higher density pyrolytic graphite matrix than is obtained when depositing from $CH_3SiCl_3$, at 3,200°F. A higher matrix density would be expected to increase the erosion resistance of the composite coating.

From the data gathered during this initial series of experiments, it was indicated that the use of $SiCl_4$ as the source will yield coatings at a significantly higher deposition temperature than can MTS. The possible advantages of this higher deposition temperature may, however, be outweighed by the poorer dispersion of the SiC phase within the PG matrix.

Eight deposition runs were conducted in the 4-inch Pereny furnace utilizing 2-inch I.D. tubular specimens, 10 inches in length. After deposition the coated tubes were sectioned transversely into five segments at 2-inch increments from the injector. These segments were then polished and examined microscopically. The coating thickness was measured microscopically and the SiC concentration was estimated visually.

Coating process data are shown in Table XXV and coating data are shown in Table XXVI.

The first deposition run was conducted at 3200°F using deposition gas flow rates established during prior codeposition studies. The first 3-1/2 inches of the substrate tube were covered with a soft deposit. The next portion of the coating was hard and gray in color. Where present, the SiC phase existed as long, stringlike crystalline spikes normal to the substrate surface. The SiC crystals were more massive than those previously grown from MTS. The dispersion was also poorer than that found when using MTS as the silicon source.

The second deposition run was conducted for 2 hours at 3200°F. During the first hour no hydrogen was used. During the second hour, hydrogen was added at twice the rate of Run No. 1. The hydrogen addition increased the SiC concentration but significantly decreased the coating deposition rate. From this observation it appears likely that the hydrogen addition inhibits the deposition of PG rather than increasing that of the SiC. The SiC phase in both portions of the coating was in the form of thick crystalline spikes with a definite tendency to cluster at the PG cone boundaries. The PG cones contained significant amounts of intraconical delaminations.

Deposition Runs 3, 4, 5 and 6 were conducted at 3600°F using various deposition gas flow rates. With the exception of deposition Run No. 4, this series yielded coatings of inferior quality, and the SiC phase showed poor dispersion where present. Run No. 4 contained moderately well dispersed SiC for the first 6 inches. The crystalline form was thick and spike like. Although the majority of the crystals were oriented 90 degrees to the substrate surface, a significant amount could be seen growing at angles up to 45 degrees from the normal plane. Visible within the coating were many pure PG growth cones which contained fine strain lines and some intraconical delaminations. Figure 22 shows the coating microstrucutre achieved in Run No. 4 at the 4-inch position.

Table XXV. Coating Process Variables Using $SiCl_4$.

| Run No. | Annulus $N_2$ (SCFH) | $N_2$ (SCFH) | $H_2$ (SCFH) | $N_2$ to Bubbler (SCFH) | $CH_4$ (SCFH) | Deposition Temperature (°F) | Deposition Time (min) | Log Book No. |
|---|---|---|---|---|---|---|---|---|
| 1 | 5.0 | 117 | 8.5 | 8.5 | 4.0 | 3200 | 60 | 289-62 |
| 2a[a] | 5.0 | 117 | 0 | 8.5 | 4.0 | 3200 | 60 | 289-62 |
| 2b[a] | 5.0 | 117 | 17.0 | 8.5 | 4.0 | 3200 | 60 | 289-62 |
| 3 | 5.0 | 117 | 17.0 | 8.5 | 4.0 | 3600 | 60 | 289-63 |
| 4 | 5.0 | 220 | 17.0 | 8.5 | 4.0 | 3600 | 60 | 289-63 |
| 5 | 5.0 | 220 | 8.5 | 8.5 | 4.0 | 3600 | 60 | 289-67 |
| 6 | 5.0 | 220 | 8.5 | 5.0 | 4.0 | 3600 | 60 | 289-67 |
| 7 | 5.0 | 220 | 17.0 | 8.5 | 4.0 | 3800 | 60 | 289-68 |
| 8[b] | 5.0 | 220 | 8.5 | 8.5 | 4.0 | 3600 | 60 | 289-68 |

[a] Two coatings applied

[b] 50/50 $SiCl_4$ and MTS mixture.

Table XXVI. Appearance of Coating at 2-Inch Increments from Injector Tip.

| Run No. | 2 inches | 4 inches | 6 inches | 8 inches | 10 inches |
|---|---|---|---|---|---|
| 1 | Covered with soft punky deposits | (0.010 in.,[a] 50%[b]) SiC visible as thick stringlike, acicular crystals. All in cone boundaries. | (0.014 in.,40%) SiC visible as long stringlike crystals surrounding patches of PG | (0.014 in.,40%) Same as at 6 in. with slightly less SiC | (None - None) Little visible coating |
| 2a | (0.009 in.,25%) SiC visible as long stringlike crystals. | (0.016 in.,15%) SiC visible as stringlike crystals concentrated at PG cone boundaries | (0.020 in.,10%) SiC all concentrated in PG cone boundaries. Many large PG cones containing delaminations. | (0.020 in.,10%) SiC visible as crystal line strings surrounding cones. | (0.010 in.5%) SiC visible as occasional irregular particles and strings at cone boundaries |
| 2b | (0.00[illegible] in.,60%) SiC visible as thick crystalline growths with patches of PG interpersed. | (0.010 in.,60%) Same as at 2-inch position. | (0.010 in.,40%) Dense aggregates of SiC are interspersed with PG cones. Coating delaminated extensively. | (0.006 in.,10%) Same as at 6-inch position. | (0.005 in.,25%) Very occasional particle of SiC. |
| 3 | (0.010 in., 0%) No visible SiC phase. | (0.018 in.,15%) SiC visible as acicular spikes moderately well dispersed. Occasional patches of PG can be seen. SiC crystals oriented at angles up to 45° of normal. | Coating very porous and rough. No measurements practical. | Same as at 6-inch position | Same as at 6-inch position |

[a]Coating thickness.
[b]Percent SiC area in microstructure (estimated).

Table XXVI. (continued).

| Run No. | 2 inches | 4 inches | 6 inches | 8 inches | 10 inches |
|---|---|---|---|---|---|
| 4 | (0.007 in., 75%) SiC visible as coral shaped growths with interspersed PG patches. | (0.011 in., 35%) SiC visible as acicular spikes and stringlike crystals. Dispersion is fair. | (0.016 in.,20%) Same as at 4-inch position. Dispersion not as good. | (0.012 in.,5%) Very occasional crystal of SiC visible. | (0.009 in.,0%) No visible SiC phase. |
| 5 | Coating soft and crumbles to touch | (0.014 in.,15%) SiC visible as acicular spikes. Tends to concentrate in PG cone boundaries. Extensive areas of pure PG are visible. | (0.017 in.,5%) SiC visible as two bands of acicular spikes, one at substrate and other near inner surface. | (0.012 in.,0%) No visible SiC phase. | (0.008 in., 0%) No visible SiC phase. |
| 6 | (0.015 in.,5%) SiC visible as an occasional spike at PG cone boundaries. | (0.007 in.,5%) Same as at 2-inch position. | (0.018 in.,0%) No visible SiC phase. | (0.013 in.,0%) No visible SiC phase. | (0.009 in.,0%) No visible SiC phase. |
| 7 | No visible SiC phase at any location. | No visible SiC phase at any location. | No visible SiC phase at any location. | No visible SiC phase at any location. | No visible SiC phase at any location. |
| 8 | No visible SiC phase at any location. | No visible SiC phase at any location. | No visible SiC phase at any location. | No visible SiC phase at any location. | No visible SiC phase at any location. |

Figure 22. Microstructure of PG/SiC Made with $SiCl_4$ - Run No. 4 (289-63) Magn. 200X.

The seventh deposition was conducted at 3800°F using the deposition gas flow rates of Run No. 4. No SiC phase was visible in any area of the coating.

The eighth and last deposition run was conducted at 3600°F using a 5-/50 MTS and $SiCl_4$ mixture as the silicon source. No SiC phase was visible in any area of the coating.

## SECTION IX

## QMS STUDIES

The quantitative metallurgical system, QMS, is capable of automatically scanning a photomicrograph of a two-phase system such as pyrographite/silicon carbide codeposit and measuring the total area, total length and breadth along two perpendicular axes, and total number of particles of one of the phases. From this, it automatically calculates the average particle area, the average length, L, and width, D, of the particles, and the elongation ratio, L/D, and the mean free path. It also is capable of automatically yielding the particle size distribution of the second phase. The microphotograph may be taken by either light optics or by electron optics. In any case, good color contrast between the phases and good edge definition of the particles must be obtained.

Three PG/SiC specimens were submitted to Micron, Inc. under this program. The following data were requested: (1) total area of field, (2) total area occupied by SiC particles, (3) total number of SiC particles in field, (4) average area of SiC particles, (5) average length of SiC particles in longitudinal direction, (6) average length of SiC particles in transverse direction, (7) average elongation ratio, (8) mean free path, and (9) SiC particle size distribution. The results of the QMS study at Micron, as well as of the pertinent prior examination at Atlantic Research, are summarized in Table XXVII. The complete QMS study is contained in the Micron reports which are attached as Appendices A and B. It was attempted to check the QMS results by comparison with: (1) approximation of the average particle area by light optics at Atlantic Research, and (2) percent SiC area calculated from the SiC concentration obtained at Atlantic Research from density measurements.

This first specimen submitted to Micron, No. 162-74, was the product from a test firing of a 1-inch subscale nozzle, CM4. The particular PG/SiC codeposit area examined was near the throat and near the substrate. The specimen was electropolished at Atlantic Research using KOH electrolyte to improve the definition of the SiC particle outline and to improve the color contrast between the SiC and the graphite matrix. The resulting microstructure is contained in Appendix A. Color contrast and phase edge sharpness are quite good. The particular area examined appears considerably coarser, at least in some parts, than the Code 21 that was assigned to this material. This explains, at least in part, why the QMS gave an average particle size area of 19.4 square microns compared with the expected area, from the code, of 4.5 square microns. The SiC area comparison in the table also suggests that the particular area examined by QMS was lower in SiC content that the entire specimen.

The second specimen, 001-52, was a portion of the coating from a 7-inch throat insert from the Scale-up Program. Micron's QMS examination is described in Appendix B and the results are compared with Atlantic Research light microscopy and composition in Table XXVII. Atlantic Research prepared and photographed the specimen by light microscopy. After diamond polishing, the specimen was etched electrolytically in 3 percent KOH at 15 volts and 5 amperes for 60 seconds. Kodak Tri-X film was given a 30-second exposure and the positives were printed on Kedalith high contrast paper. Micron repolished and then photographed the same specimen using electron imaging techniques. As seen in Figures 1 and 2 of Appendix B, the resolution with the electron imaging was quite poor. The average particle area agreement with Atlantic Research was best in the case of the light optics, as seen in Table XXVII. On the other hand, the percent SiC area showed best correlation in the case of the electron microprobe. Micron did not conduct the particle size distribution analysis because of the poor resolution obtained. It should be noted that Micron had confused Specimen No. 4, previously submitted and studied in Appendix A, with Run 001-52.

## Table XXVII. QMS Summary.

| Codeposit Run No. | ARC Micro Code | ARC Weight Percent SiC | Average Particle Area ($\mu^2$) | | SiC Area (%) | | QMS L/D | Specimen Parent Shape |
|---|---|---|---|---|---|---|---|---|
| | | | QMS | ARC[a] | QMS | ARC[b] | | |
| 162-74 CM-4 0332-12 | 21 | 27 | Optical 19.4 | 4.5 | Optical 11.8 | 20 | 1.5 | 1-inch throat insert |
| 001-52 6334-1 | 21 | 29 | Optical 3.8 | 3.1 | Optical 15 | 21 | Optical 1.9 | 7-inch throat insert |
| | | | Electron Probe 6.5 | | Electron Probe 23 | | Electron Probe 2.4 | |
| | | | SEM 7.5 | | SEM 30 | | SEM 2.1 | |
| 033-16 | (20) | (24) | SEM 2.3 | (4.5) | SEM 27 | (17) | SEM 1.2 | 1-inch throat insert |

[a] Average width squared x QMS elongation ratio.

[b] Based on weight percent SiC.

The third specimen studied at Micron was PG/SiC graded codeposit from a 1-inch throat insert, 033-16. The QMS results with this material are also reported in Appendix B and compared with Atlantic Research results in Table XXVII. This was a graded coating and the QMS work was done on the PG/SiC area near the substrate. Consequently, the Atlantic Research data reported in Table XXVII are an average of the data from the 1-inch throat inserts that were fired under the Air Launch Program. This particular specimen was examined by the scanning electron microscope, SEM, only, in the unpolished condition; i.e., in the as-coated condition. Fair checks with Atlantic Research data were obtained. The SEM photographs of this specimen, which are shown in Appendix B, are excellent (see Figure 4 of Appendix B) and show some gradation in SiC crystal population from the substrate (bottom of Figure 4) outward. However, it is doubtful whether edge definition is adequate with SEM since the flat surface is not photographed. The lower than expected elongation ratios obtained in this study may be explained by the fact that the long needles appear dominant to an observer of a microstructure whereas the small nonacicular particles are generally not too noticeable.

This brief study should be regarded as preliminary. It does demonstrate the QMS technique may have potential. It's use would appear to be more feasible after improved reproducibility and homogeneity are achieved. At least until that time, ten or twelve areas should be examined and the results averaged.

One area in which the current QMS technique appears to be lacking is that of defining the degree of dispersion of the particles. The mean free path is only an average of the distance between the particles. An attempt should be made to define the degree of distribution of the free path about this mean. SiC particles often segregate in and/or have a higher elongation ratio in the PG grain boundaries.

A number of specimens have been submitted to AFML and Micron, Inc., under the SCALE-UP Program. Twelve areas of each specimen are being examined by electron optics. Some tentative results have been received orally from Lieutenant Hollenberg at AFML. This indicates good agreement between the SiC by QMS and Atlantic Research density determinations, provided the SiC crystals are not finer than Code 20. AFML also find that the best edge definition and phase contrast can be obtained with the electron microprobe. They are also attempting to measure the degree of dispersion of the particles.

A specification on the QMS, as applied to PG/SiC codeposit, will be issued at a later date as a separate document.

## SECTION X

## DISCUSSION

A summary of the major coating run variables is listed in Table XXVIII. The most common microstructure found with these test fired coatings was code 20 but some codes 10, 11, 21, and 30 were also considered to be acceptable. No clear pattern for the microstructural variation emerges although it is thought to be associated mainly with deposition temperature, thermal gradient and gas flow rate. Composition and deposition rate were never considered to be under good control, although they were known to be functions of temperature, MTS content of the reactant gas, and probably residence time and temperature gradient. The molar ratio of $CH_4$/MTS in the reactant gases is approximately 7. The deposition reactions are:

$$CH_4 = C + 2H_2$$

and

$$CH_3SiCl_3 = SiC + 3HCl$$

Assuming stoichiometry, the PG/SiC ratio in the product should be 7. However, it is approximately 14. This indicates that pyrolysis, of the MTS, is not complete. This effect is even more pronounced at higher temperatures.

Although the effect of deposition temperature was not determined systematically, in this series, it has been and is known that an increase in temperature coarsens the SiC needles and decreases the SiC content. In this work, every attempt was made to hold the temperature at 3200°F. Frequent checks of the optical pyrometers using primary temperature standards as well as checks against other recently calibrated instruments showed the pyrometers to always be in calibration to within 50°F.

The distance between the injector tip and the substrate was determined to be extremely important, particularly in controlling the coating thickness profile. The 1.0-inch throat insert entrance to throat thickness ratio was quite inconsistant until the spacing was increased by 0.5 inch. It then remained at 0.75±2 for the remainder of the 1.0 inch series. Similarly, the same thickness ratio for the 1.7 inch HIPPO inserts became consistent only after run 26 when the spacing was increased by 1 inch.

Another coating variable that may be important is the thermal gradient along the furnace axis. This may explain why coatings change in character when a new furnace element is installed.

The nitrogen flow rate is also considered to be important in that the PG cones are smaller and the coating surface is smoother at higher flow rates.

A few special points on good coating technique were discovered in this investigation. At least a 15 minute soak time at temperature is required and, further, the MTS must be turned on prior to the methane presumably in order to provide sites for growth of the SiC needles. Otherwise, the SiC crystals may be coarse and the dispersion poor early in the deposition cycle. Another point of good technique is that the substrate must be clean of all loose particles and debris prior to coating.

Table XXVIII. Coating Run Variables Summary
(for coatings that were test fired only).

| | Percent $CH_4$ | Percent MTS | Temperature (°F) | Deposition Rate (mils/hr) | | | Percent SiC | | Micro Code |
|---|---|---|---|---|---|---|---|---|---|
| | | | | Entrance | Throat | Exit | Entrance | Exit | |
| 1.0-inch Throat Inserts | 1.4 | 0.22 | 3200 | 12 | 16 | 14 | 27 | 21 | 21 |
| 1.0-inch Entrance Approach Sections | 2.8 | 0.40 | 3200 | 6/24 | -- | 25 | 15/22 | 15/34 | 11/20 |
| 1.7-inch Throat Inserts | 1.6/2.4 | 0.23/0.38 | 3200 | 17 | 20 | 19 | 20 | 21 | 20 |
| 1.7-inch Entrance Approach Sections | 1.5/2.9 | 0.12/0.46 | 3200/3500 | 12/24 | -- | 14/32 | 12/25 | 15/47 | 11/30 |

The dimensional changes on the outside envelope of the components change only very little during coating. It appears, from this work, that the change is detectable only on sufficiently large components; in this case the 1.7-inch HIPPO entrance approach sections.

The number of coating runs made in each type of furnace and for each type of coated component are listed in Table XXIX. The Phase I Task 1 goal was achieved in that sufficient pieces for the 1.0-inch subscale (7) and the 1.7-inch HIPPO tests firings (8) were fabricated. However, it was not possible to control the coating parameters so as to accurately engineer the effect of a change in substrate geometry or size. Preliminary coating experiments must still be conducted with each new geometry. A fundamental coating study to assess the effect of all coating parameters is recommended.

Table XXIX. Phase I - Task 1 Coating Runs.

| | 4-inch Pereny Resistance Furnace | 6-inch Pereny Resistance Furnace | 6-inch Induction Furnace | Total |
|---|---|---|---|---|
| 1.0-inch Throat Inserts | 28 | 0 | 0 | 28 |
| 1.0-inch Entrance Approach Sections | 7 | 5 | 0 | 12 |
| 1.7-inch Throat Inserts | 38 | 2 | 4 | 44 |
| 1.7-inch Entrance Approach Sections | 0 | 11 | 16 | 27 |
| Totals | 73 | 18 | 20 | 111 |